

**INVESTIGATION INTO OBSTACLES TO IMPLEMENTATION OF THE
DIRECTED SELF-ASSEMBLY OF BLOCK COPOLYMERS**

A Dissertation
Presented to
The Academic Faculty

By

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In Partial Fulfillment
of the Requirements for the Degree
Doctor of Philosophy in the
School of School of Chemical and Biomolecular Engineering

Georgia Institute of Technology

May 2018

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DIRECTED SELF-ASSEMBLY OF BLOCK COPOLYMERS**

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Date Approved: March
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”Three and Nine”

ACKNOWLEDGEMENTS

I would like to start off by thanking Dr. Clifford Henderson and Dr. Pete Ludovice for advising me throughout my graduate studies. Dr. Henderson has given me direction whenever I've needed it with my research and been a great source of new ideas for current and future projects. Dr. Ludovice brought a refreshing dose of humor to our group meetings as well as been very helpful whenever I hit a wall with my simulations. I would also like to thank the remaining professors who served on my thesis committee including Dr. Carson Meredith, Dr. Zhiqun Lin, and Dr. David Bucknall.

I'd like to thank the many now-graduated group members who helped me get on the right track with my research from the beginning. Dr. Richard Lawson was an excellent post-doc when I joined the group, excellent at challenging ideas from multiple angles. Dr. Andrew Peters gave me my first real taste of molecular dynamics simulations, showing me the ropes of how to use our computer cluster, as well as re-introducing me to pokemon games (an excellent game for down time). For my first year in the lab Dr. Jose Baltazar was very supportive in helping me think through my early experiments. In my first year as well Dr. Nathan Jarnagin taught me how to conduct polymer synthesis and over the years has been great company when talking about new potentially useful block copolymers. Arriving in the group at roughly the same time as I did, Brandon Sharp and Hannah Narcross have also been good companions along this journey. Both chemists are a good source of humor and have aided me whenever I needed help discerning a reaction mechanism or troubleshooting a reaction. I'd like to also thank Haibo Li. He and I together conducted three months of grueling long-hours lab work to get the anionic polymerization synthesis procedure shown in this work to where it is.

My friend and colleague Dr. Benjamin Nation arrived at Georgia Tech the same time as I did along with our good friend Dr. Krishna Jaychandrababu. The three of us went on multiple outings throughout the course of my graduate studies for various foods, bowling,

and movies. Ben joined the Henderson group at the same time I did and while he was more geared towards simulation work he offered great insight into my lab experiments as well. I had the honor of being one of Ben's groomsman at his wedding, a celebration I was very grateful to have been a part of. Ben taught me the more intricate aspects of the model used for his and my simulations. He was always a good competitor at the games we played whether it be pokemon or chess, the latter of which he demolished me in and the former we were evenly matched.

Lastly, but certainly not least, I'd like to thank my family members for their support over these years. The family has grown quite a bit in such a short time with three marriages and three extra kids. I'd probably be nothing but skin and bones were it not for my mom's care packages, full with the best fuel for research, candy. My family over these years have been excellent getaways from lab work full of fun and games during the winter breaks or the Birthday Bash. I'm sure the family and fun will only continue to grow in the future, and I'm grateful for it all.

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List of Abbreviations

AFM	Atomic force microscopy
BCP	Block Copolymer
DO	Defect Order
DSA	Directed Self-Assembly
EUV	Extreme Ultraviolet
χ	Flory-Huggins Interaction Parameter
ΔF	Difference in Free Energy
GPC	Gel Permeation Chromatography
GPU	Graphics Processing Unit
ITRS	International Technology Roadmap for Semiconductors
L_0	Spacing (Pitch or Domain) of a BCP
LER	Line Edge Roughness
LWR	Line Width Roughness
N	Degree of Polymerization
n_{Homo}	Fraction of chains that are homopolymer
NA	Numerical Aperture
NMR	Nuclear Magnetic Resonance
ODT	Order Disorder Transition
PSP	Pinning stripe position
PS	Polystyrene
PMMA	Poly(methyl methacrylate)
PHEMA	Poly(2-hydroxyethyl methacrylate)
PtBS	Poly(4-tertbutylstyrene)
PPMA	Poly(propyl methacrylate)
q^*	Position of the primary scattering peak

SAXS	Small angle x-ray scattering
SEM	Scanning electron Microscope
TBDMS	Tert-butyl dimethyl silyl protecting group
T_g	Glass Transition Temperature
ϕ_i, f_i	Volume fraction of component i

SUMMARY

To meet the growing demands of the microelectronics industry and their desire to continue Moore's Law, a variety of routes to extend or replace optical lithography have been suggested. Among these options is the directed self-assembly (DSA) of block copolymers (BCPs) which have the ability to microphase separate into features with spacings as low as sub-10 nm. The DSA of these features are typically achieved by graphoepitaxy or chemoepitaxy which use either topography in the substrate or chemically preferential pre-patterns in the substrate, respectively, to direct the BCP's phase separation. Despite the use of these techniques, BCPs suffer from several roadblocks to their implementation in chip manufacturing including production of adequate BCP materials, high line edge roughness (LER) and line width roughness (LWR), and high defect densities. This work explores possible reasons for high LER, LWR, and defects using coarse-grained molecular dynamics and expands the library of BCP materials by synthesizing two new BCPs.

Chapter 3 uses simulations to investigate the effect pinning stripe position, density multiplication, and defect order on the relative free energy of dislocation defects for BCP films on chemoepitaxial underlayers. In Chapter 4 the effect of homopolymer concentration on LER and LWR is explored for BCP/homopolymer blends. Chapter 5 and 6 show the synthesis and characterization of Poly(4-tertbutyl styrene)-block-Poly(propyl methacrylate) (PtBS-b-PPMA) and PtBS-b-Poly(2-hydroxyethyl methacrylate) (PtBS-b-PHEMA), respectively. PtBS-b-PPMA is a new low χ BCP that may be useful in applications such as photonic crystals and filtration membranes as well as in discerning the relationship between defect annihilation rates and χ . PtBS-b-PHEMA is a new high χ BCP that has shown via SAXS profiles to be able to phase separate into features with a sub-7 nm pitch.

CHAPTER 1

INTRODUCTION AND BACKGROUND

This chapter is intended to give an introduction to block copolymers (BCPs) and directed self-assembly (DSA). First a brief history of optical lithography will be discussed. Following this a detailed explanation of BCPs and their ability to self-assemble will be described. Next the primary methods for directed self-assembly for BCP thin films will be detailed. Finally an overview of major challenges facing implementation of BCP-DSA will motivate the proceeding chapters.

1.1 Optical Lithography

It is self evident that over the past few decades there has been a boom in the technological advancement of electronics. In 1971 the first commercially available microprocessor was made (the Intel 4004) with about 2000 transistors and a minimum feature size of 10 microns.[1] The transistor count of a chip, roughly speaking, gives a measure of the chips functionality, meaning the higher the transistor count, the more functions can be performed. For this reason, the semiconductor and microelectronics industry's goal, starting with the Intel 4004, has been to increase the number of transistors that they can fit in a given area on a chip. To do so, it means the spacing between features on a chip needs to decrease as well. For this reason, the minimum feature size and/or the transistor count are used to mark advancements in chip manufacturing. In the mid 1960s an economic trend, coined Moore's Law (Figure 1.1), was noted by Gordon Moore who predicted that roughly every two years the number of transistors on a chip will double.[2] This has been the guideline for the industry since the 70s, with every company in the industry attempting to meet this prediction. As a result computers that once filled an entire room can now fit in a person's pocket and possess far more functionality. As stated before, the microprocessors used for

this technology had roughly 2300 ($2.3 \cdot 10^3$) transistor and a minimum feature size of 10 microns, while more modern chips have transistor counts of multi-billions ($> 10^{10}$) and feature sizes as low as 10 nanometers (0.01 microns).

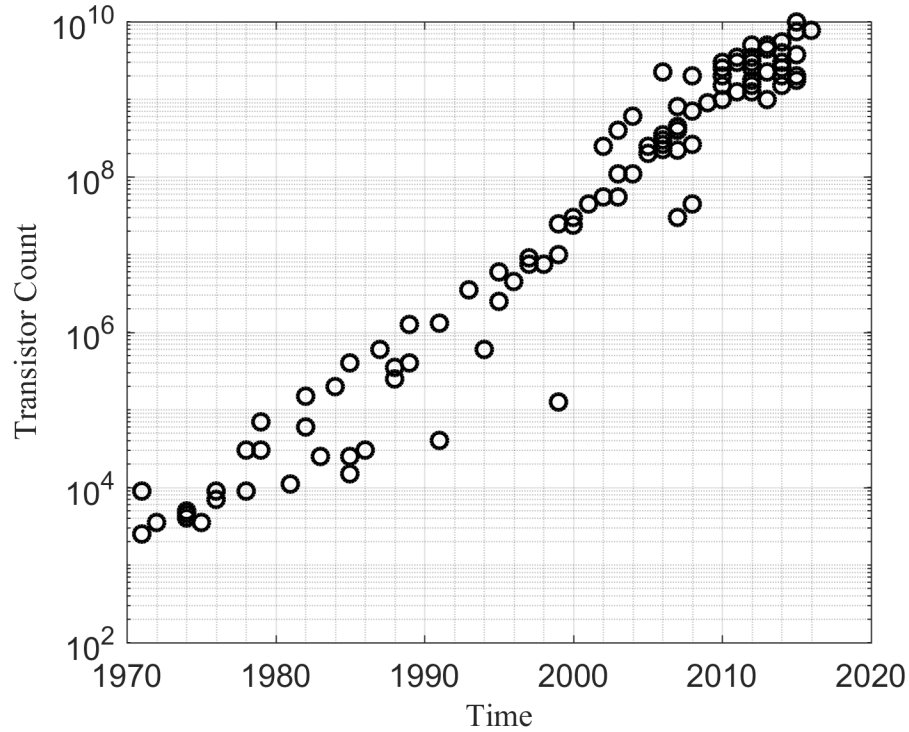


Figure 1.1: Moore's law is depicted here by the increase in transistor count, roughly doubling for a given chip size every two years.

To make such a high density of features, the microelectronics industry for the most part has relied on a process known as optical lithography. A brief overview of the process is shown in Figure 1.2. Initially a thin film of a photoresist is coated onto a substrate (Figure 1.2A). Light of a particular wavelength (λ) is then shined at a chrome mask that has holes in either the shape of the features desired or the inverse of said features (Figure 1.2B). Depending on the type of photoresist employed, the exposed regions (Figure 1.2C) are either crosslinked and made insoluble (negative tone, Figure 1.2E) or chain scission occurs and they are made soluble (positive tone, Figure 1.2D) compared to the unexposed regions. The photoresist is then developed, a process where the more soluble regions are removed

by immersing the wafer in a solution. The photoresist is then developed by using a solution to wash away the more soluble regions, leaving behind a mask on the substrate. The silicon substrate is etched or doped in the unprotected regions and then the mask is removed.

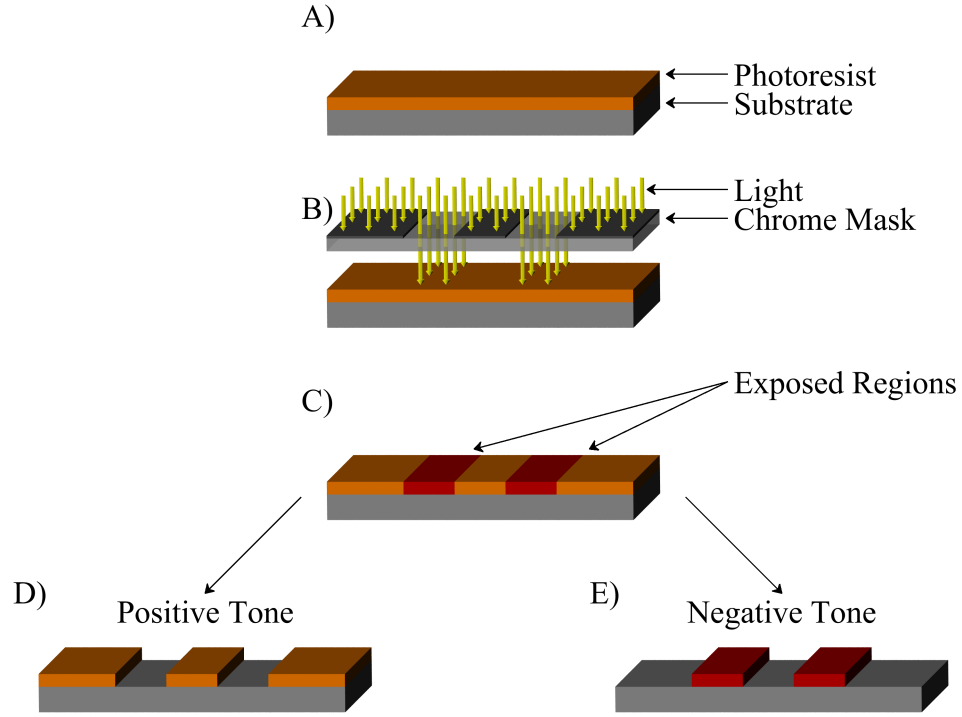


Figure 1.2: Illustration of general procedure for optical lithography.

Optical lithography is limited by both the minimum feature size the process can resolve and the depth of focus. Equation 1.1 and 1.2 demonstrate how the wavelength of light (λ), the index of refraction of the medium through which the light travels (n), the angle between the lens projecting the light and the photoresist (θ), and process parameters (k_1 and k_2) affect both the resolution (R) and the depth of focus (DoF).

$$R = \frac{k_1 \cdot \lambda}{n \cdot \sin(\theta)} \quad (1.1)$$

$$DoF = \frac{k_2 \cdot \lambda}{(n \cdot \sin(\theta))^2} \quad (1.2)$$

The product $n \cdot \sin(\theta)$ is also called the 'numerical aperture' (NA). Typically, in order to decrease the minimum feature size that can be resolved, λ is either lowered or the NA is increased. However, by doing so, the depth of focus decreases as well, making it more challenging to pattern uniform features through the full thickness of the photoresist. The process parameter k_1 is specific to minute details in the lithographic setup that may change from lab to lab, but is expected to have a theoretical limit of 0.25 as its lowest value.[3] Much of the driving force for lowering feature size has been by decreasing the wavelength of the exposure light. When λ decreases however, many aspects of optical lithography need to be changed as well, such as the type of photoresist used, the dosage required, lens construction, and so forth.[4, 5, 6]

Originally, light sources for exposure tools was based on the spectral lines of Hg-lamps (436 nm, 405 nm, and 365 nm).[7] Modern exposure tools use light sources with either 248 nm or 193 nm wavelength light produced from excimer lasers.[7] Nearing the end of the lifespan of 193 nm lithography, 157 nm lithography was expected to take its place. However, due to complications with finding proper lens materials, among other things, 157 nm lithography never came to fruition.[7, 8] This led to the industry implementing a series of "tricks" to increase the lifespan of 193 nm lithography with the reduction of feature sizes while further research into the successor of 193 nm lithography continued. Among these "tricks" included techniques such as immersion lithography and self-aligned double patterning. As stated previously, by reducing NA , the minimum resolvable feature size decreases. This can be achieved by changing the index of refraction (n) of the medium through which the exposure light travels (Equation 1.1). Immersion lithography achieved this by placing a thin layer of water between the mask and the photoresist, reducing feature sizes, and dubbing the process as 193-i.[9, 10] Self-aligned double patterning on the other hand is a method in which patterns are made in a photoresist, etch transferred to a sacrificial layer, and a new material known as a "spacer" is then grown onto the sacrificial layer.[11, 12] The spacers have a pitch equal to the feature size of the patterned sacrificial

layer, and upon etching of the sacrificial layer, the feature density of the spacers is twice that of the feature density originally patterned by the photoresist. However self-aligned double patterning increases the number of layers on a wafer and the number of steps in chip manufacturing, increasing cost.

As the demand for smaller and smaller feature sizes increases however, the pressure is on the microelectronics industry to uphold Moore's Law. Several technologies are being considered for implementation to either replace or extend optical lithography. Among these include electron beam lithography, nanoimprinting, advancements in immersion lithography, extending double patterning, extreme ultraviolet lithography (EUVL), and use of block copolymer directed self assembly. These alternatives will be discussed briefly. Extending double patterning to quadruple patterning and higher is unattractive due to the increase in wafer layers and manufacturing steps required for pitch reduction.[13] While 193-nm lithography was a success, finding liquids with higher indices of refraction that will not interfere with properties of the photoresist has been challenging.[14] Electron beam lithography (E-beam lithography) can pattern small feature sizes by using one or more e-beams to pattern photoresists directly, but even with multiple e-beams this would be a very slow process.[15, 16] Nanoimprinting uses a pre-patterned mold placed over a thin film to form features that will be used as a mask for the substrate..[17, 18] Upon heating, the fluid fills in the spaces of the mold. The fluid is then cured, the mask lifted to be reused, and the pattern transferred to the substrate. However defects in the mold, slow throughput, and lift-off issues with the mold prevent nanoimprint's commercial use.[17, 18]

EUVL is one of the more promising routes to increasing feature density.[19, 20, 21] This process would reduce λ from its current 193 nm to 13.5 nm, allowing for sub-10 nm feature size patterning. At 13.5 nm wavelength, instead of a chrome mask to block the light, special molybdenum-silicon (Mo-Si) mirrors must be made to reflect unwanted light. For a typical EUVL tool multiple Mo-Si mirrors are needed to send the light from source to wafer, leading to a large reduction in light intensity by the time the light reaches the

photoresist. This means long exposure times are expected. To produce the light for the EUVL tool, a tin droplet is excited by a CO₂ laser and photons are then emitted at 13.5 nm wavelength. This light is then captured by a large Mo-Si mirror surrounding the droplet and directed down the path of the previously mentioned series of mirrors to the wafer. During this process, debris from the tin droplet can damage the mirror, leading to a decrease in the amount of captured light from the source. While these challenges and more hinder EUVL's implementation, the industry is devoting a large amount of time and effort to addressing them.

Along side EUVL, directed self assembling block copolymers are a leading contender in the extension of optical lithography. Block copolymers (BCPs) are known to phase separate into periodic features with a pitch (feature to feature distance) of less than 100 nm.[22, 23, 24, 25] The goal of using BCPs here is to allow thin film BCPs to phase separate into guided periodic features over a substrate, remove one of the blocks, and then use the remaining BCP as a direct mask with which to etch/dope the substrate (Figure 1.3). The following sections will go into detail describing what a block copolymer is, how directed self assembly is implemented, and the current challenges with its implementation.

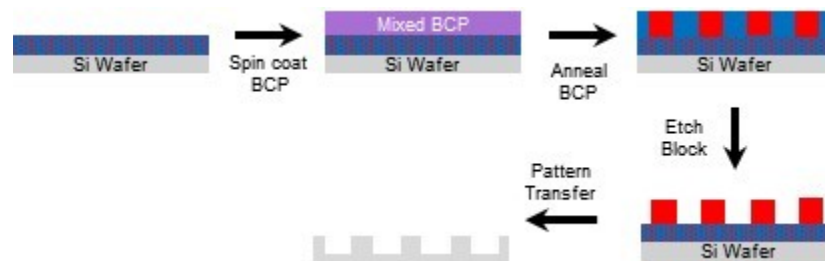


Figure 1.3: The intended use of BCPs for the microelectronics industry is to use the ordered BCP morphology to create a mask for direct patterning of features into the substrate.

1.2 Block Copolymers

A polymer is a long chain composed of repeating units called "mers", hence its name poly-mer (many-mers). A polymer with a single type of repeat unit is called a homopoly-

mer. Were two homopolymers connected by a single covalent bond at their chain ends, they would be deemed an A-B linear diblock copolymer (Figure 1.4a). Other forms of block copolymers exist as well, for example if three homopolymers were joined by covalent bonds an A-B-C or A-B-A triblock copolymer can be formed. However the focus of this work is diblock copolymers and this will be used interchangeably with “block copolymer” from this point forward unless otherwise stated.

Block copolymers (BCPs) have a unique ability to microphase separate into a variety of morphologies (Figure 1.4b). If two containers of oil and water are combined, they do not mix and form two phases completely separated from one another. If oil and water are replaced by two homopolymers, a similar effect may occur. As the two homopolymers attempt to reduce the number of interactions between each other, small pockets of one homopolymer may form inside the other. Block copolymers wish to do the same, however they are prevented by the covalent bond connecting their blocks. Due to this, the block copolymer may microphase separate forming periodic local regions with high concentration of one block and then another (Figure 1.4b).[22, 23, 24, 25]

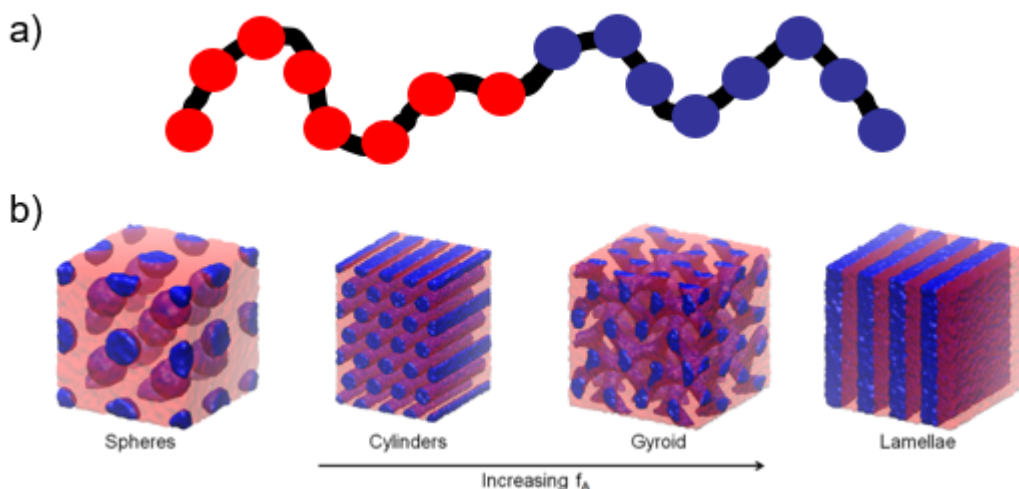


Figure 1.4: Depiction of a diblock copolymer (a) as two homopolymers linked by a covalent bond. Due to this constraint, BCPs can phase separate into different morphologies by varying the volume fraction (f_A) of each block in the BCP (b).

The morphology of the phase separated BCP is dictated by the relative volume fraction (ϕ or f_i) of the blocks in the BCP, and the product χN . [26, 27] Here, N is the degree of polymerization of the BCP (how many mers are in the chain). The parameter χ is known as the Flory-Huggins parameter which describes the degree to which one block prefers to interact with itself rather than the other block. [28] The driving force for the BCP to mix is entropy, preferring that there be a lack of any order in the system. However, the product χN is a measure of the enthalpic penalty for unlike blocks interacting with one another, and is thus the driving force for microphase separation in a BCP. A BCP's morphology will be dictated by the relative space filling ability of each block (ϕ) and the reduction of surface area to decrease unlike block interactions. Together, these two competing forces act to drive down the free energy and stabilize the BCP system (Equation 1.3).

$$\Delta G_m = R \cdot T \cdot (n_1 \cdot \ln(\phi_1) + n_2 \cdot \ln(\phi_2) + n_1 \cdot \phi_2 \cdot \chi_{12}) \quad (1.3)$$

Equation 1.3 is known as the Flory-Huggins equation where, n_i is the number of moles of component i , ΔG_m is the Gibbs free energy of mixing, R is the universal gas constant, and T is the temperature of the system. Based on the minimization of free energy, a self-consistent mean-field bulk phase diagram for an extremely ideal bulk BCP was crafted (Figure 1.5a).[26, 29] The ideal BCP used in these calculations was symmetric in density, symmetric in cohesive energy density, symmetric in statistical segment length, and monodisperse. From Figure 1.5a however it can be seen that if a BCP is too short (low N) or its blocks have a low penalty for mixing (low χ), microphase separation will not occur and chains will disorder. Similarly, if one block is very small in comparison to another (very low ϕ_i), the BCP will disorder as well. The point at which microphase separation begins to occur is known as the order-disorder transition (ODT). For a 50/50 volume fraction ideal BCP, this occurs at $\chi N = 10.5$. Above the ODT, morphologies predicted by mean field theory (with increasing ϕ_i) were body centered cubic spheres, hexagonally closed packed cylinders, and lamellae. After experimental verification, later calculations included

a gyroid phase at lower values of χN between the cylindrical and lamellar phases.[29, 30] Another phase found experimentally, was the perforated lamellae phase which is disputed as being kinetically trapped and at non-equilibrium.[31, 29, 32] While Figure 1.5a shows a bulk phase diagram for an ideal BCP, real bulk BCP phase diagrams (Figure 1.5b) are not quite as symmetric in appearance and may differ in the position of the phase boundaries or ODT.[27]

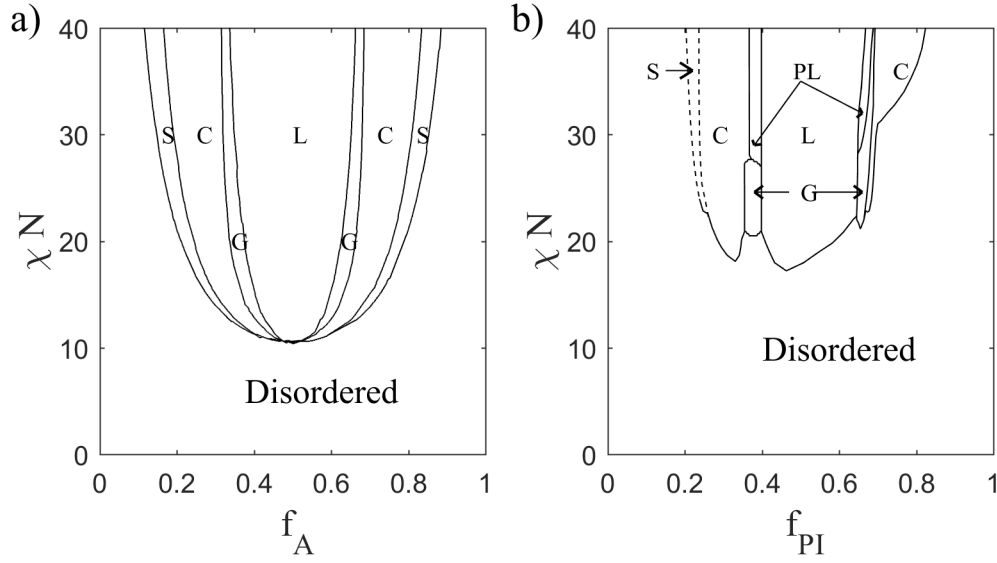


Figure 1.5: Mean field phase diagram (a) for an ideal BCP displays symmetry in the windows of its phases. However real BCPs are not nearly as symmetric in density, cohesive energy density, or conformation, leading to more asymmetric phase diagrams such as the one for PS-b-PI [27] (b).

For BCPs there exists three regions known as the strong segregation regime (SSR), weak segregation regime (WSR) and the intermediate segregation regime (ISR). These differ primarily by their scaling of pitch (L_o) to N (Equation 1.4) due to the degree of chain stretching that occurs.[26, 29, 33, 34]

$$L_o \approx a_{BCP} \cdot \chi^{\frac{1}{6}} \cdot N^{\alpha} \quad (1.4)$$

Here, a_{BCP} is the statistical segment length of the BCP and α is the parameter that

varies depending on which regime you are in. In Equation 1.4 it should be noted that while L_o is dependent on both χ and N , it is more strongly dependent on N . This leads to a trade off for BCP systems. In order to form morphologies with a small feature size ($\frac{1}{2} \cdot L_o$) a BCP with a low N will be required. However, by lowering N there is a risk of dropping below the ODT and the BCP being unable to microphase separate. Therefore, a BCP with an inherently high χ is needed such that even at low N s the BCP will still remain above the ODT. Up to this point fundamental concepts about BCPs in the bulk has been discussed. However for the application in Figure 1.3 a thin film of the BCP will be needed. The next section discusses the self-assembly of thin film BCPs and how their two interfaces (substrate and free-surface) can effect their utility.

1.3 Block Copolymer Thin Film Directed Self-Assembly

BCPs are of interest to the semiconductor and microelectronics industry primarily due to their ability to form lamellae and hexagonally closed pack (HCP) cylinders. Lamellae offer a means to pattern alternating line-space patterns while HCP cylinders can be used for patterning contact holes.[35, 36, 37, 38] Other applications for BCPs include areas such a filtration membranes,[39, 40, 41] drug delivery,[42, 43, 44] and photonic crystals.[45, 46, 47] For membrane filtration a matrix composed of HCP cylinders with the ability to control their diameter can be useful for filtration by size exclusion. Drug delivery usually employs micelle forming BCPs with a hydrophilic shell and a hydrophobic core where the core acts as a carrier for the drug in question. Photonic crystals are a class of material with periodic structures on the scale close to that of natural light ($L_o > 100nm$). At these pitches, particular wavelengths of light may be reflected or trapped. This leads to photonic crystals being of interest for applications such as anti-reflective coatings, fiber optics, and additives for pigments. Due to their periodic natures and driving force to form defect free arrays, BCPs find potential utility as photonic crystals. For the majority of these applications, BCPs are initially prepared as thin films on a substrate.

Before a BCP is coated onto a substrate, a suitable underlayer must first be chosen. If a 50/50 lamellae forming BCP is used, after phase separation the lamellae may be present perpendicular (Figure 1.6a) or parallel (Figure 1.6b) to the substrate. Two primary factors affect which orientation will occur for the BCP film: surface energy and film thickness. If the substrate the BCP is coated on has a high preference for one block, or the free surface has a high preference for one block, that block will want to wet the surface and parallel orientation will be preferred.[48, 49] In extreme cases when a block prefers to interact with the free surface while the BCP is on a neutral underlayer, a wetting layer may form at the free surface, covering perpendicularly oriented features.[50] If a BCP film is a thickness that is an integer multiple of the half pitch ($0.5L_o$) then the chains of the BCP are able to nicely stack perpendicular to the substrate (forming parallel features). This is known as having a film thickness that is commensurate with the pitch. If the film thickness however is not an integer multiple to the half pitch (incommensurate), then perpendicular features will be preferred due to the chains inability to stretch or compress enough to form parallel features.[51, 52] For lithographic applications, generally the phase separated features are desired to be oriented perpendicular to the substrate.

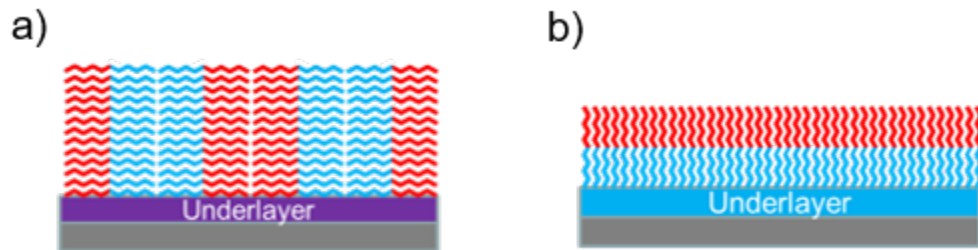


Figure 1.6: Perpendicular orientation (a) can be achieved when the substrate and free surface are neutral and do not prefer to interact with one block over another. Parallel orientation (b) occurs when either the free surface or substrate has a preferential interaction with one of the blocks.

Were a lamellae forming BCP coated as a thin film on a neutral substrate (the substrate interacts with each block equally), it may initially be in the disordered state or phase sepa-

rated with a high density of defects. In order to phase separate and/or reduce the number of defects in the BCP, their chains need mobility and this is typically achieved in one of two methods. The act of going from a disordered or defective morphology to a well ordered morphology in the BCP film is known as “annealing”. The first is known as “thermal annealing” where by the temperature of the thin film is heated above the glass transition temperature (T_g) of the BCP.[48] Above this temperature polymers exhibit increased mobility and diffusion while below this temperature they are relatively low mobility. Above the T_g the BCP chains will phase separate and order, however if the temperature is raised too high the risk of degradation becomes present.

For BCPs whose individual blocks have high T_g s, the degradation temperature kinetically limits their annealing and phase separation takes more time. For these specific cases however the second method of increasing chain kinetics is used, known as “solvent annealing”.[53, 54, 55] Solvent annealing operates by allowing the BCP film to swell with a particular solvent and effectively lower the T_g to below the current film temperature. Unlike thermal annealing however, solvent annealing may inadvertently change the effective volume fractions of the blocks. If the penetrating solvent equally partitions into both blocks of the BCP, the inherent block volume fractions remain unchanged. However if a preferential solvent is used, one block may swell more than another leading to horizontal shift along the phase diagram and possibly a different morphology may be reached than what the inherent BCP would attain.[55, 56, 57] As solvent concentration increases [58, 59, 60] or film temperature increases [61, 62, 63, 22], the effective χ of the BCP decreases as well. In some cases, a combination of the two methods (solvo-thermal annealing) may be necessary to achieve adequate chain mobility.[64]

After annealing a lamellae forming BCP film on a neutral substrate, a top down view would show lamellae traveling in multiple directions. This is known as a “fingerprint lamellae” due to the lack of guidance or direction of the lamellae (Figure 1.7). Fingerprint lamellae do not offer much utility for patterning since the direction of their features are

not controlled. In order for BCPs to be useful, a means of guiding their phase separated features is necessary. The guidance of the phase separating BCP's features is known as "directed self-assembly" (DSA) and can be primarily achieved using either graphoepitaxy or chemoepitaxy. While graphoepitaxy guides the BCP via topography in the substrate, chemoepitaxy uses pre-patterns in the substrate that have a high preference for interacting with one of the blocks in the BCP.

In graphoepitaxy (Figure 1.8a), a thin film is first coated onto a substrate and patterned, forming a large trench.[65, 66, 67, 68, 69, 70, 71] The BCP is then coated, partially or fully filling the trench. The material of trench is preferential to one of the blocks in the BCP and the block in question wets the sidewall of the trench it resides in. This forces the BCP chains to orient parallel to the substrate, leading to perpendicular features. The preferential sidewall is a strong guiding force for the BCP, and therefore the phase separated lamellae will follow parallel to this sidewall. The primary factors affecting graphoepitaxy are the surface energy of the sidewall, the surface energy of the substrate, and the width of the trench.[65, 67] If the trench is commensurate with L_o perpendicular features are likely to form while if the trench is incommensurate with L_o parallel features are more likely to form. However, if the trench is commensurate, but the sidewall is not adequately preferential to one of the blocks, a "ladder" morphology of perpendicular lamellae will form.[65]. It is therefore essential that both the trench be close to commensurate with L_o and the sidewall be preferential to one of the blocks in order to properly guide the BCP lamellae.

In chemoepitaxy (Figure 1.8b) the guiding force for the lamellae come from chemical preferences in the underlayer.[72, 73, 74, 75, 76, 77] An underlayer is constructed that has two regions, a "pinning" stripe and a "background region". The pinning stripe is highly preferential to one of the blocks in the BCP while the background region is neutral or has a slight preference for the other block in the BCP. The level of guidance is dependent on the chemical contrast between the pinning stripe and background region and the relative widths of these regions. An oversized or undersized pinning stripe may lead to footing

or undercutting in the lamellae's through-film profile, which will affect its efficacy as a mask.[78, 79] Typically the background region is much larger than the pinning stripe, and thus if its surface energy is too preferential it can lead to defects or even flipping of the orientation of lamellae over it.[80, 81]



Figure 1.7: Illustration of a top-down view of a phase separated lamellae forming BCP on only a neutral underlayer.

In chemoepitaxy or graphoepitaxy were a pinning stripe placed every $1L_o$ or the width of the guiding trench $1L_o$ there would be a high penalty for the lamellae to do anything but follow the pre-pattern guiding it. However if the technology was available to make such small periodic patterns then BCPs would not be needed at all. One of the major benefits of BCPs towards this application is that even if there are some missing or defective pre-patterns in the underlayer, the BCP may still be properly guided.[76, 77] This benefit is taken advantage of by instead of making the width of the trench (graphoepitaxy) or spacing from pinning stripe to pinning stripe (chemoeptiaxy) exactly $1L_o$ it is made an integer multiple of L_o . The integer multiple is known as the “density multiplication”. This means for example a chip manufacturer could pattern an underlayer with 14 nm line-space patterns using conventional optical lithography, and then by using DSA-BCPs a mask with 7 nm (2x density multiplication) line-space patterns could be produced. However, the higher the density multiplication the lower the guiding force for the BCPs. An example

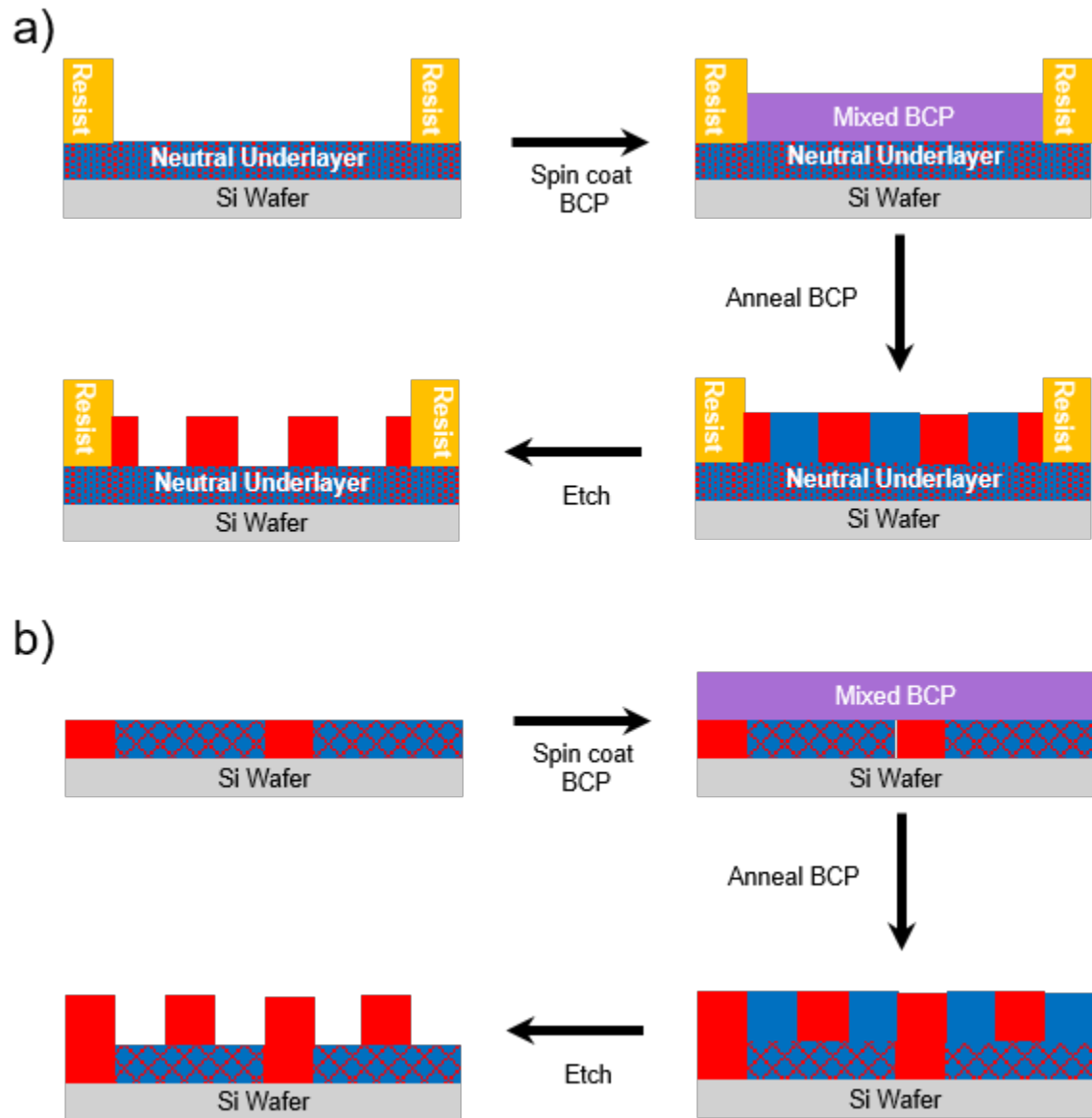


Figure 1.8: In graphoepitaxy (a) the driving force for guidance is by the sidewall of the pre-patterned resist (yellow). In chemoepitaxy (b) the driving force for guidance is the energetic contrast between the pinning stripe and the background region in the underlayer.

of a working chemoepitaxial process is the Lui-Nealey process (LiNe process) which uses a crosslinked random copolymer as a background region and a grafted homopolymer as the pinning stripe.[72] More sophisticated processes for DSA use hybrids of grapho- and chemoepitaxy such as the SMART [82] and COOL [83] processes.

1.4 Challenges in the Directed Self Assembly of Block Copolymers

As with the other alternatives or enhancements to optical lithography, BCP-DSA faces its own set of challenges. The International Technology Roadmap for Semiconductors (ITRS) monitors and tracks the progression of lithographic techniques or alternatives, setting standards for various aspects of chip production and determining when a method is ready for industry implementation. Three challenges facing implementation of BCP-DSA in the microelectronics industry are in 1) production of proper BCPs, 2) defects in the phase separated features, and 3) the line edge roughness (LER) and line width roughness (LWR) of the features.[84, 85]

An optimal BCP would be one that has a high enough χ to allow for low features sizes when phase separated, one that anneals in a short period of time, one that does not require a topcoat, and one that has a relatively high etch contrast between its two blocks. One of the most common BCPs used in research today is polystyrene-block-polymethyl methacrylate (PS-b-PMMA). PS-b-PMMA is synthesized easily by most polymerization methods and both blocks have a T_g of around 100°C. This higher than room-temperature T_g allows for the phase separated features and BCP chains to be "frozen" in place at room temperature. The measured χ of PS-b-PMMA is between 0.03 and 0.04 and temperature has minimal effect on the value.[61] After annealing however PS-b-PMMA has a useful (good pattern transfer) limit of about a 22 nm pitch between features leading to a desire for BCPs with a higher χ that can reach smaller useful feature sizes.[86] PS-b-polyisoprene (PS-b-PI) was found to have a $\chi = 0.06$ at 150°C.[87] PS-b-PI is also one of the handful of BCPs that have a well mapped phase diagram (Figure 1.5b).[27] Other notable high- χ BCPs include PS-

b-Polydimethylsiloxane (PS-b-PDMS, $\chi = 0.30$ at 150°C) [88], Polycyclohexylethylene-b-PMMA (PCHE-b-PMMA, $\chi = 0.18$ at 150°C) [22], and PS-block-Polyhydroxystyrene (PS-b-PHOST, $\chi = 0.12$ at 150°C) [63].

With ever increasing χ , current progress has led to materials able to produce sub-10 nm pitch morphologies, but extension of this library of BCPs is still necessary. As χ increases so does the difference in cohesive energy density of the BCP's blocks which may lead to one block preferentially interacting with one of the thin film's surfaces and forming a wetting layer.[50] For instance, while PS-b-PDMS may have the highest χ in the previously mentioned BCPs, it also has a propensity to form a wetting layer of PDMS at the free surface which would require pre-etching before it can be used as a lithographic template. If a wetting layer is likely to form at the free surface, a neutral top coat may be required to ensure the microdomains align perpendicularly to the substrate.[89] This top coat is undesirable as its coating and removal increase the number of steps industry has to perform and lowers throughput. For this reason increasing the library of high χ BCPs is necessary to find a BCP that has a high χ that a neutral underlayer can be produced for, but that does not require a top coat.

While having a high χ is necessary to decrease the L_o between BCP microdomains, it may also lead to difficulty in defect removal.[90] From the initial application of an underlayer, to the phase separation of the BCP, and even during the transfer of the pattern to the substrate, defects have the potential to occur. A distinction needs to be made though between defects caused by the phase separation of the BCP and defects caused by non-BCP related issues. For instance, if a particle lands on the underlayer prior to the BCP being coated, this will affect the resulting alignment of the BCP's features.[91] However, if the BCP is phase separating on a patterned underlayer, and its microdomains are unable to follow the pre-pattern, then a BCP defect may occur (Figure 1.9). Defects in the BCP are akin to line defects in crystals in that dislocations, disclinations, or jogs in the lamellae may occur.[92] These defects are expected to be unfavorable due to the higher surface area

and interfacial interactions they incur to the BCP that would otherwise not be there in a defect-free case.

The ITRS has set a defect density target for BCP-DSA of allowing 1 defect every 100 cm².^[93] Due to the difficulty of tracking defects insitu, much of the work on understanding how defects annihilate has been done using computer simulations. The following chapter will discuss these types of simulations in further detail. Of the defects previously mentioned, dislocations like the one in Figure 1.9 have been the main focus for initial studies. The gray dots shown in Figure 1.9 represent what will be called the "terminating blocks" of the defect. When defect annihilation occurs, there is a fluctuation of the concentration profile at these terminating blocks, followed by a bridging from their domain, across the unlike block, and to the adjacent domain (Figure 1.10B and C).^[90] Once this bridging occurs, molecular diffusion then annihilates the defect (Figure 1.10D and E). The latter parts of this process (molecular diffusion) has a set timescale dependent on factors such as N and the temperature of the film. However the initial bridging happens spontaneously, and is the rate limiting step to defect annihilation.^[90] During this process there is an increase in the number of interactions between the two blocks (as compared to the original defective state) and for high χ materials this can be highly unfavorable. This means that although there is expected to be a high penalty for the defect to be present, there is an expected higher energetic barrier preventing it from reaching a defect-free state. It is expected that a low χ material may lead to increased defect annihilation kinetics, however; low χ materials rarely see as much attention as high χ materials. Expansion of the library of low χ BCPs will be useful in determining the trade offs in χ and defect kinetics.

Simulation work thus far has focused on predicting the likelihood for a defect to occur by measuring its relative free energy to a defect-free state.^[92] By using a Boltzmann's distribution (Equation 1.5) the relative free energy can be used to give a probability for a defect to occur. In Equation 1.5 ΔF is the relative free energy between a defect and defect-free state, k_B is the Boltzmann's constant, T is the temperature, and ρ_D is the probability



Figure 1.9: Example of a Dislocation defect in a top-down image of phase separated lamellae forming BCP. The gray dots denote the 'terminating block' of the defect.

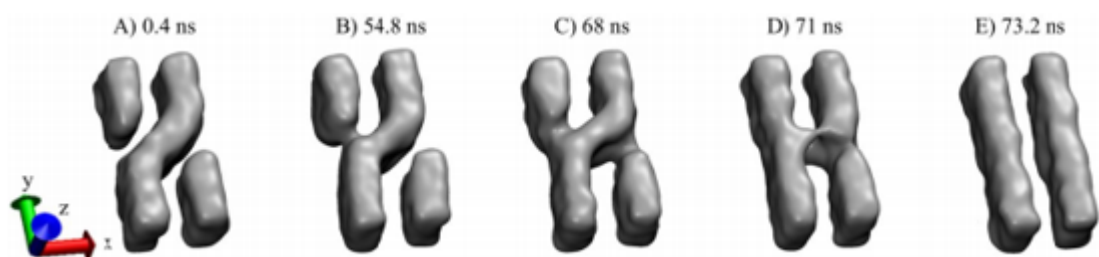


Figure 1.10: Illustration of dislocation defect annihilation (A). Initially bridge formation between the two terminating blocks (B and C). Molecular diffusion then occurs moving the BCP from the defective region until it is annihilated (D and E)[90]

a defect will occur. Thus far all results have given very high ΔF values (> 200 kcal/mol) giving very low probabilities for defects. For this reason it is suspected that defects are kinetically trapped by the high energetic penalty for their annihilation as mentioned earlier. However, simulations have only probed very specific BCP systems with very ideal simulated BCPs. Further simulation work into a wider variety of BCP-DSA systems (different underlayers, block copolymer compositions, or more realistic block copolymers) would be of benefit.

$$\rho_D \approx \exp\left(\frac{-\Delta F}{k_B \cdot T}\right) \quad (1.5)$$

Line edge roughness (LER) and line width roughness (LWR) refers to the error in the position of the BCP interface and the error in the consistency of the feature size ($0.5L_o$). High levels of roughness in patterned features can affect the operation of transistors and be detrimental to the performance of integrated circuits.[94, 95] Because of this, low levels of roughness are tolerated, for instance at the 14 nm node the acceptable LWR was only 1.1 nm.[93] For bulk BCPs the roughness of the interface is primarily controlled by the degree of segregation (χN), with large χN BCPs having a sharp interface (low LER) and low values of χN typically having broad interfaces.

However for a thin film BCP increased roughness could be caused by factors such as the patterned underlayer or the thickness of the film. Assuming a perfectly patterned chemoepitaxial underlayer, as the film thickness increases the lamellae closer to the free-surface is less influenced by the patterns in the underlayer and can deviate from that pattern placement without incurring a heavy penalty. For the lamellae near the patterned substrate however it has been shown that factors such as the width of the pinning stripe and the composition of the background region can also influence the roughness.[78] It has also been shown that the polydispersity (PDI) of the BCP being used can affect the roughness for PDIs > 1.3 .[96] The PDI is a measure of the distribution of chain lengths in a given BCP sample, with a PDI = 1 being monodisperse (all the chains have the exact same length).

However it is unclear how additives such as homopolymer can affect both the LER and LWR. Adding homopolymer to a BCP is an attractive method for modulating the pitch of the phase separated features given the ease at which homopolymer can be synthesized compared to synthesizing an exact molecular weight BCP for a particular L_o . [72]

The work presented in this thesis is split into two parts: simulation (Chapters 3 and 4) and experimental (Chapters 5 and 6). In Chapter 2 the model used for the coarse grained molecular dynamics simulations and general analysis procedures for those simulations will be discussed. Chapter 3 will discuss the effects of density multiplication, pinning stripe position, and defect order has on the relative free energy of a dislocation defect compared to a defect free state for BCP thin films on chemoepitaxial underlayers. In Chapter 4 the effect of homopolymer addition and pinning stripe width has on LER and LWR will be explored for chemoepitaxy underlayers with 2x density multiplication. The experimental work of this thesis primarily deals with the synthesis and characterization of new block copolymers. Chapter 5 details the synthesis and self-assembly of a new low χ BCP poly(4-tertbutyl styrene)-block-poly(propyl methacrylate) (PtBS-b-PPMA) while Chapter 6 details the synthesis and self-assembly of a new high χ BCP poly(4-tertbutyl styrene)-block-poly(2-hydroxyethyl methacrylate) (PtBS-b-PHEMA).

CHAPTER 2

MODEL AND ANALYSIS METHODS

In this chapter the model used for the molecular dynamics simulations in Chapter 3 and 4 will be discussed. The first section will discuss what a molecular dynamics model is and the particular model used for this work. The next section will detail how the initial states of the simulations were built for both bulk and thin film simulations. Then how simulations were actually started and run from their initial states will be discussed. Finally, the last section will detail the typical methods for analysis of simulations by producing gray scale images and measuring their line edge- and line width roughness.

2.1 Coarse Grained Molecular Dynamics Model

Molecular dynamics models essentially work on the principle of Newton's second law of motion. Objects in the simulation are given an initial state, forces act upon them over a particular timestep, and these forces can be summed to determine the acceleration of the object. The force (F) on an object in the simulation can be determined by the negative gradient of the potentials (E) applied to the object (Equation 2.1).

$$F = -\nabla E \tag{2.1}$$

The acceleration can then be determined by Newton's second law, stating that the force on an object is equal to the product of its mass and acceleration. If the current position and velocity are known, the acceleration can be used to determine the new position and velocities at the next timestep. The new positions and velocities can then be used to calculate new accelerations and the cycle can repeat until the simulation has run the specified number total of timesteps. A molecular dynamics simulation must keep track of the po-

sitions, velocities, and accelerations of every atom in a simulation. The time required to run a simulation will increase when more atoms are added to the simulation. Likewise, the simulation time will also increase with the number of timesteps. In order to simulate phenomena that occur on relatively large size and time scales, such as BCP microphase separation and annealing, the polymers are coarse-grained. In coarse-graining, multiple atoms are combined into one unified “bead”, referred to as beads for the rest of this work, as a method of decreasing the number of particles the simulation must track. Additionally, by coarse-graining the simulation is able to take larger timesteps since many of the high frequency atomic phenomena are now ignored, such as atomic bond vibrations. Figure 2.1 shows an illustration a typical BCP chain in our simulations where each bead represents four repeat units of the BCP chain.

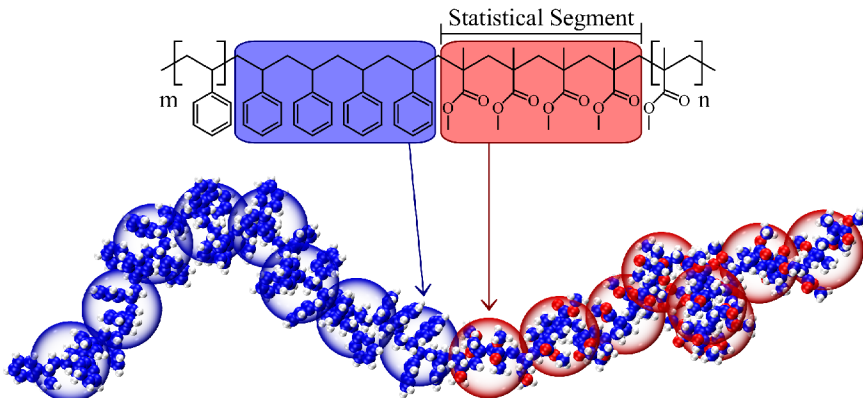


Figure 2.1: Illustration of coarse graining for a typical BCP chain in simulation. Here, four monomers are represented as a single, unified bead - roughly the statistical segment length of PMMA. The BCP used in this work can be considered a slightly more ideal chain than PS-*b*-PMMA.

The BCP used in this work is parameterized to be ideal, that is, symmetric in properties between the red beads and blue beads. The persistence length of a BCP chain describes how long you can travel along the chain before it changes direction. This “length” can also be described in “number of repeat units”. For PMMA there is a persistence length of

roughly four repeat units, and this was chosen as the coarse graining for the BCPs in this model.[90]

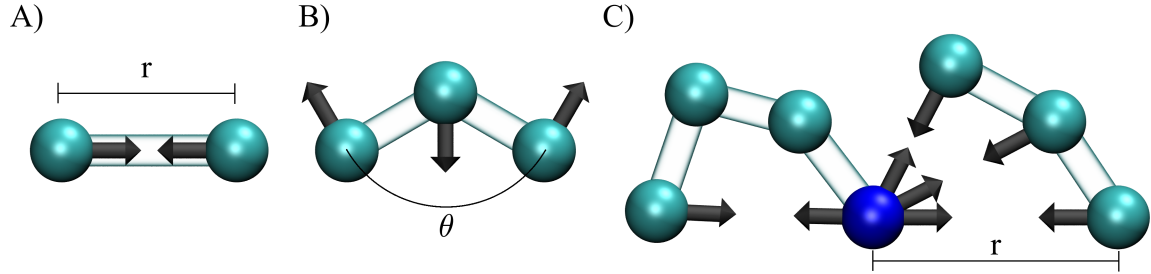


Figure 2.2: Illustration of the bond potential (A), angle potential (B) and non-bonded potential (C) felt by beads in simulations. Gray arrows depict the force vectors. For the non-bonded potential the arrows are acting upon the blue bead.

In a typical simulation, there are three types of potentials: a stretching potential, an angle potential and a non-bonded potential. These potentials are illustrated in Figure 2.2 and described by Equations 2.2 - 2.4. The stretching potential (E_{str}) is used to bond a series of beads together and simulate the polymer chain's bonding along its backbone.

$$E_{str}(r) = k_{bond} \cdot (r_{eq} - r)^2 \quad (2.2)$$

In Equation 2.2, k_{bond} is the bond force constant, r_{eq} is the equilibrium bond length, and r is the current length of the bond. For all work in this thesis $k_{bond} = 100$ kcal/(mol nm²) and $r_{eq} = 0.82$ nm. Along a chain, every set of three beads, feels an angle potential described by Equation 2.3. This angle potential was used to ensure the chain behaves more like a real polymer.

$$E_{ang}(\theta) = k_{ang} \cdot (\theta_{eq} - \theta)^2 \quad (2.3)$$

In Equation 2.3, E_{ang} is the angle potential, k_{ang} is the angle force constant, θ_{eq} is the equilibrium angle between a set of three beads, and θ is the current angle between the set of three beads. For all the work in this thesis $k_{ang} = 5$ kcal/(mol rad²) and $\theta_{eq} = \frac{2}{3} \pi$. A

non-bonded potential (Equation 2.4) is used to simulate the cohesion of the homopolymers and a χ between beads along a given chain and between beads on different chains.

$$E_{ij}(r) = \epsilon_{ij} \cdot \left[\frac{\sigma_{ij}^8}{r} - 2 \cdot \frac{\sigma_{ij}^4}{r} \right] \quad (2.4)$$

Here, E_{ij} is the non-bonded potential felt by bead i from interacting with bead j , ϵ_{ij} is the strength of the interaction between bead i and bead j , σ_{ij} is equilibrium distance between the two beads, and r is their current distance from one another. Equation 2.4 takes the form of a lennard jones potential, but with an 8-4 exponent instead of 12-6. Effectively this broader potential increases the width of the energetic well in the lennard jones plot, and is found to work well for coarse grained systems. For all work in this thesis $\sigma_{AA} = \sigma_{BB} = \sigma_{AB} = 1.26$ nm and $\epsilon_{AA} = \epsilon_{BB} = 0.5$ kcal/mol. The value of ϵ_{AB} may vary depending on the simulation. Since ϵ_{AB} describes the strength of the non-bonded potential between unlike beads, it has a relation to χ for a BCP (Equation 2.5).

$$\chi = z \cdot \left[\frac{1}{2} \cdot (\epsilon_{AA} + \epsilon_{BB}) - \epsilon_{AB} \right] \quad (2.5)$$

Where in Equation 2.5, z is the coordination number given by Flory and acts as a constant in these simulations.[28, 97] The value of z is determined by running a simulation of a disordered BCP at various ϵ_{AB} , calculating the scattering profile, and then fitting Leibler's theory to the primary peak to determine χ . Equation 2.5 is then fit to these χ values and z is determined.[90] For all the work in this thesis z was determined to have a value of 4.3. For simulations in this thesis, ϵ_{AB} varies from 0.5 to 0.35, where ϵ_{AB} of 0.5 gives a χ of 0 and ϵ_{AB} of 0.35 gives a χ of 0.547. All of the BCP chains in this work have an $N = 64$ with 16-beads (8 of each block), giving a $\chi N = 35$.

Molecular dynamics simulations are run using HOOMD-Blue on an in-house GPU cluster consisting of a mix of Nvidia GTX 580, GTX 680, and Titan Black enabled systems [98, 99] as well as the CIRCE cluster at the University of South Florida [100]. Simulation

results were viewed using MATLAB [101] or Visual Molecular Dynamics (VMD) [102] utilizing the Tachyon ray tracing library.[103]

2.2 Initial States

Simulations built are split into either bulk BCP simulations with periodic boundaries in the x , y , and z directions, or thin film simulations with a finite z dimension and periodic x and y dimension. To build a bulk simulation first the number of beads, desired size of chains, and dimensions for the simulation box are used to determine the number of chains capable of fitting in the simulation. To generate a polymer chain two beads are placed at the equilibrium bond distance (r_{eq}) apart from another. One after another the remaining beads are added such that every two beads are at a distance of r_{eq} and each set of three beads are at the equilibrium angle (θ_{eq}) until the chain is the appropriate length. The completed chain is then placed randomly within the simulation box. Due to the random packing of the chains, it is possible for two beads from different chains to overlap one another leading to a massive repulsive force. This is avoided by two minimization procedures which will be described in the next section.

For thin simulations (periodic in the x and y and finite in z), BCP thin films interact with an underlayer. The underlayer and the BCP are made separately, with the BCP film being made similar to bulk simulations. A brush underlayer is made by creating a short six bead chain, $N = 28$, with the same parameters as the BCP (though labeled different for easier identification). The chains are then "grafted" to a substrate by fixing all of their end beads in space at a set z position. The grafted bead ends are positioned along a grid in this z plane with a surface density of 0.44 chains/nm². To prevent the non-grafted chain end from inverting away from the direction of the soon to be BCP film, a layer of fixed beads are placed beneath the grafted chain ends at the same surface density but offset to the previous grid's position. This second grid helps to more accurately simulate a hard surface. The underlayer is then placed just below the BCP film.

Generally when these films are built, the chains of the BCP film are randomly placed into the simulation box, giving the appearance of a disordered mixture of the BCP chains. However sometimes the films are built "pre-aligned" in orientations similar to their phase separated state. To achieve this, BCP chains are rotated such that the primary axis of the radius of gyration tensor is placed along the x dimension, the secondary axis is placed along the y dimension, and the tertiary axis is placed along the z dimension. The chain's center of mass is then placed along where a lamellae interface is expected to be. When all the chains are placed into the box, the appearance of a small gap can be seen in the middle of where the BCP lamellae would be present. This gap is filled once the simulations are allowed to start running. Figure 2.3 shows examples of the initially mixed and initially pre-aligned simulation states for thin film and bulk simulations.

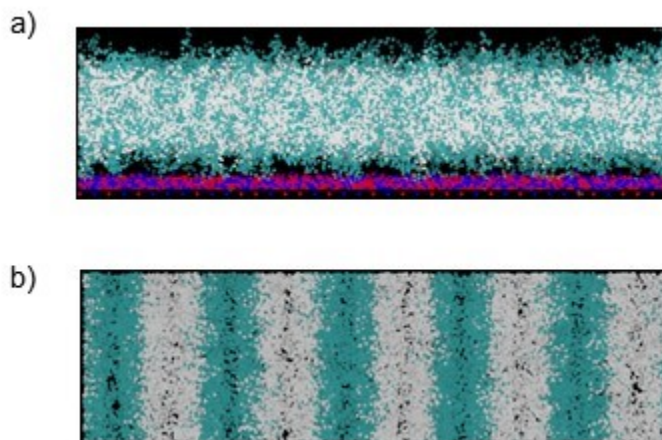


Figure 2.3: Examples of the initial build states of thin film NVT simulations (a) and pre-aligned bulk NPT simulations (b). The black areas are vacuum in the simulation box.

For BCP-homopolymer blend studies, homopolymer is made from pre-existing BCP chains. To generate homopolymer in an A-B BCP film, first the film is made as previously stated. Next, chains are broken apart into their constitutive homopolymers until the desired homopolymer loading is present. To break apart a chain, the bond between the 8th and 9th beads must be removed, as well as the angle potential between 7-8-9 and 8-9-10. Homopolymer chains have the same properties and parameters as their A-B counterparts

including the same ϵ_{ij} .

2.3 Typical Simulations

As stated previously the initial state may have some beads very close to one another in position. Were the simulation started at this point there would be a high repulsive force between these beads and may cause the simulation to crash. Therefore two minimization steps are needed to steadily move beads apart so that this excessive repulsion can be avoided. The first minimization gently pushes all beads apart by setting σ_{ij} to a low value ($\sigma_{ij} = 0.001\text{nm}$), reducing the repulsive force beads close together would feel. Next a series of minimization steps, 50 steps each are run, increasing σ_{ij} by 0.005nm at the start of each new run until σ_{ij} reaches 1.26nm . This minimization is run using HOOMD FIRE (Fast Inertial Relaxation Engine [104]) minimizer (parameters: $dt = 5 \times 10^{-6}$, $ftol = 1 \times 10^{-2}$, $Etol = 1 \times 10^{-7}$, $finc = 1.99$, $fdec = 0.8$, $\alpha_{start} = 0.01$, and $\alpha = 0.9$).

The second minimization is then run at a longer time interval (20,000 steps). During this minimization the beads are allowed to feel repulsive forces for a small amount of time, letting them increase their distance from one another steadily until their separation is near r_{eq} . The FIRE minimizers [104] is used here (parameters: $dt = 5 \times 10^{-4}$, $ftol = 1 \times 10^{-2}$, and $Etol = 1 \times 10^{-7}$). After the minimizations, generally a 200,000 timestep (10 ns) period is given for the chains to interact while $\epsilon_{AA} = \epsilon_{BB} = \epsilon_{AB} = 0.5$. During this time period, there is no χ between the BCP blocks. Also during this time period, for BCP thin films, the film is allowed to collapse down onto the underlayer. (Figure 2.4a). For pre-aligned films however, this step is skipped because if there is no χ between the blocks, the pre-aligned lamellar domains will mix.

After dynamics start for a thin film simulation, the BCP film settles down on the underlayer, the chains take their natural state, and the surface becomes relatively smoother (Figure 2.4a) than in the initial state (Figure 2.3). For pre-aligned films, during the first nanoseconds of simulation time the chains take on their natural shape and fill the space in

the middle of the lamellar domains (Figure 2.4b). For thin film pre-aligned simulations, this space is then taken out of the total film thickness, slightly decreasing it from the expected film thickness.

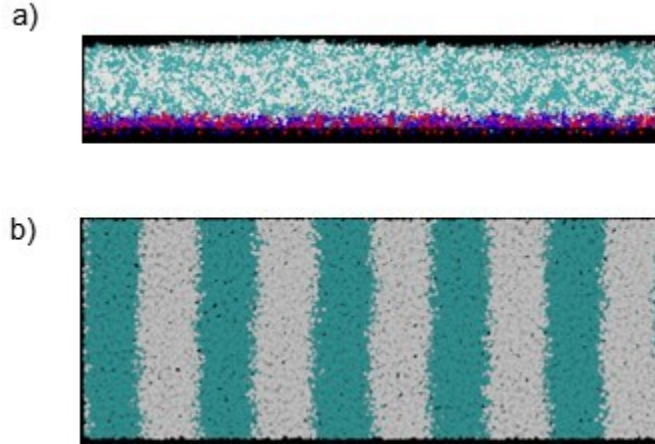


Figure 2.4: Examples of the initial build states in Figure 2.3 after 1 nanosecond with dynamics turned on in the simulation. a) is a thin film simulation while b) is a bulk simulation.

At this point, the proper simulation is started with the desired parameters. From this point forward ϵ_{AB} is set to 0.35. Initial velocities are assigned randomly, following a Maxwell-Boltzmann distribution for an initial temperature of $T = 500K$. For the thin film simulations seen in this work a HOOMD NVT integrator [105, 106] is used while for the bulk films mentioned in this thesis a HOOMD NPT integrator [105, 107, 108] is used. In both cases the integration timestep is 0.05 picoseconds. In NVT simulations the number of beads, volume, and temperature are kept constant. A Nose-Hoover thermostat is used with a temperature set point at $T = 500K$ and a coupling constant of $\tau = 0.2$ timesteps. In NPT simulations however the number of beads, pressure, and temperature are kept constant. This allows for the volume of the simulation to fluctuate. An MTK barostat-thermostat is used for NPT simulations with a temperature set point of $T = 500K$, a temperature coupling constant of $\tau = 2$ timesteps, a pressure setpoint at $P = 1atm$ and a pressure coupling constant of $\tau_P = 20$ timesteps.

NPT simulations are useful for determining the natural pitch of a BCP or BCP/homopolymer blend. Pre-aligned lamellae are placed in bulk NPT simulation with their chain's center of mass placed along where lamellae interfaces are expected to be. The lamellae are parallel to the yz plane and perpendicular to the x dimension. The y and z dimension are coupled to one another such that if there is any change in one dimension, the other changes equally, while the x dimension is allowed to freely change. Bulk simulations are typically $5 \cdot L_{est}$ in the x dimension and 20 nm in the y and z dimension, where L_{est} is an estimate for the pitch of the BCP/homopolymer blend. After minimization, once dynamics begins and χ is on, the x dimension will the stretched chains will take their natural state and the x dimension will either expand or shrink to accommodate the BCP chains. By averaging the x dimension over the last few nanoseconds of the simulation, and dividing by 5 (the number of pre-aligned repeat distances) the natural pitch of the BCP or BCP/homopolymer blend can be measured. This method is found to give equivalent repeat distances to those calculated in previous works by simulating the scattering of the BCP to calculate the pitch.[90]

2.4 Simulation Analysis

2.4.1 Generating Images

While VMD is useful at looking at individual films, MATLAB was used for generating mass-quantities of film images. Using MATLAB, an approximation to a gray scale scanning electron microscope (SEM) images could be created. In these images white regions are considered areas of high concentration of one block type, and black areas are high concentrations of the opposite block. To do this for top-down images a grid is made over the two visible dimensions (for top-down this means x and y) with points separated by 0.1 nm. At each grid point the number of each bead type within a vicinity of a 1 nm radius is counted through the entire depth of the film. In the case of only looking at the BCP film, the fraction of A-beads in that cylindrical cut of the film will be between 0 (black) and 1 (white), creating a grayscale image from the collection of grid points. An example of these

grayscale images produced by MATLAB are shown in Figure 2.5. This process is repeated for cross-sectional images with the exception that the y dimension is now being averaged through. In some cases the entire y or z dimension is not averaged through, sometimes only the bottom or 5 nm of the film are averaged over to determine what areas near those interfaces look like separately from the film.

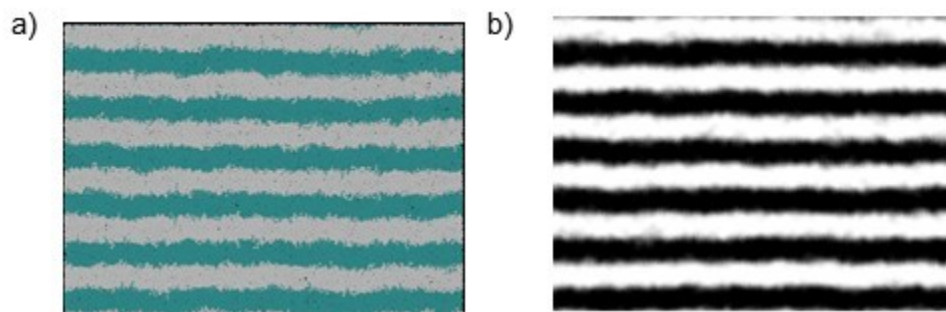


Figure 2.5: Example of simulations visualized top down in VMD (a) and their z -averaged gray scale image (b) made via MATLAB. The white beads in a) are analogous to the white parts of the image in b).

2.4.2 Calculating Roughness

Line edge roughness (LER) and line width roughness (LWR) are calculated from gray scale images of the simulations. The LWR (Figure 2.6c) is the standard deviation of the width of a lamellae as you travel along its direction, while the LER (Figure 2.6a & b) is the standard deviation of the position of a single interface of a lamellae. For a given point in time of the simulation, a contour map is made showing points along the gray scale image where the fraction of A-beads (f_A) is 0.5, an identifier for the edge of two lamellae. This contour map is made by looking at a particular grid point in the gray scale image and its nearest neighbors and trying to interpolate to an f_A value of 0.5. By doing so the points along the contour ensure that one of the position coordinates for the interface is an integer. Next the points along each interface are collected and sorted to keep track of which lamellar interface (pinned or unpinned by the underlayer) the points came from. Due to the grid

size, the points of the interface are evenly spaced by 0.1 nm. For an individual interface, the standard deviation (σ_{LER}) of the x position of the interfacial points is determined and the LER is reported as $3\sigma_{LER}$.

LER can increase by two mechanisms. The first is a roughness that occurs over a short length scale due to local variations in placement of the center of a BCP chain (Figure 2.6a). However, LER can increase over large length scales as shown in Figure 2.6b, where whole lamellae oscillate or wobble. In this case LER can increase heavily while LWR remains relatively constant, and when this occurs the lamellae interfaces are said to be “correlated”. The LWR is easily determined once the points of the interface are known. Since in the interfacial points the y coordinate is always an integer, by appropriately pairing two interfaces of a lamellae and subtracting their x coordinate, the width of the line can be determined. The standard deviation (σ_{LWR}) of those line widths is then calculated and the LWR is reported as $3\sigma_{LWR}$. Figure 2.6c gives an example of a feature with high LWR with interfaces that are completely uncorrelated.

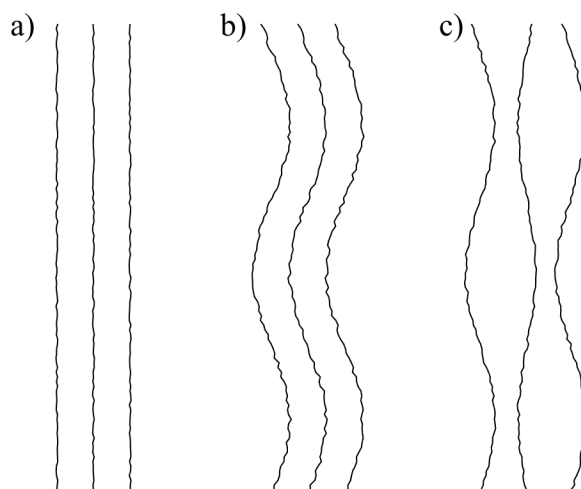


Figure 2.6: Short range fluctuations in the lamellae interface are always present, causing an increase in LER (a). However long range fluctuations can occur leading to an increase in LER while having a low LWR (b). LWR increases when the interfaces go from being “correlated” (a & b) to “uncorrelated” (c).

CHAPTER 3

FREE ENERGY OF DEFECTS IN CHEMOEPITAXIAL BLOCK COPOLYMER DIRECTED SELF-ASSEMBLY: EFFECT OF PATTERN DENSITY AND DEFECT POSITION

3.1 Introduction

For lamellae forming block copolymers (BCPs), even under directed self-assembly via chemoepitaxial underlayers, defects in the lamellae can occur. These defects take an appearance similar to line defects such as dislocations, jogs, and disclinations.[92] The ITRS has stated that the tolerance for such defects are 1 in 100 cm². [93] While determining the likelihood of a defect forming experimentally is difficult, it is an ideal question to be answered by simulations. By using a Boltzmann's distribution the defect density (ρ_D) can be related to the relative free energy of adefective state in lamellae.

$$\rho_D \approx \exp\left(\frac{-\Delta F}{k_B \cdot T}\right) \quad (3.1)$$

In Equation 3.1 ΔF_D is the difference in free energy between a defect and defect-free state, T is the temperature, and k_B is Boltzmann's constant. Previous simulation work has determined that for dislocations, jogs, and disclinations, ΔF_D is $> 300 k_B T$. [92, 90] This means that from Equation 3.1 it would be incredibly unlikely for defects to occur, leading researches to the idea that defects are merely kinetically trapped states that need longer annealing times to reach a defect-free state. However previous work on relative defect free energy was done for small dislocation defects with a limited variety of underlayers. Here, a deeper look into the behavior of defect free energy is investigated for five differently "sized" dislocation defects on chemoepitaxially patterned underlayers with 1x, 2x, 4x, and 8x density multiplication. In addition, the fundamental effect of pinning stripe position

relative to the defect is explored. By varying these conditions, the effect of underlayer pattern density, defect “size”, and pinning stripe placement on the defect free energy will be determined.

3.2 Modeling Approach

3.2.1 Model and Build

The molecular dynamics model used for this work is the same as described in Chapter 2. Thin film BCP simulations were run on chemoepitaxial underlayers that were patterned based on bead position and bead type. The dimensions of the BCP films were $8L_o \times 6L_o \times 0.75L_o$, where $L_o = 11.8631$ nm for this BCP ($N = 64$, 16-bead BCP). This allowed for underlayers with various density multiplications such as 1x, 2x, 4x, and 8x. The pinning stripes for all density multiplications were set at $0.5L_o$ while the background region is set at $(m - 0.5)L_o$ wide where m is the density multiplication. The background region was always composed of $f_A = 0.5$, where f_A is the number fraction of A-beads in the background region. To ensure the pinning stripe is highly preferential to one bead type, it is composed with an $f_{A/B} = 1.0$, depending on which block needs to be pinned.

Figure 3.1A and 3.1C show an example of films with varying density multiplication, 4x and 8x, respectively. Along with varying density multiplication the position of the pinning stripe was varied as well in relation to the defect. This can be seen in Figure 3.1A and 3.1B where the pinning stripe is in the middle of the defect (3.1A) or one lamellae adjacent from the center (3.1B). The pinning stripe position (PSP) was varied 1-lamellae position at a time until the pinning stripe straddles the periodic boundary. Due to symmetry, this means for an $8L_o$ wide simulation, there are 9 unique pinning stripe positions. In the next section the method for relative free energy calculations (thermodynamic integration) will be discussed as well as how the “size” (defect order, DO) of the defect will be varied.

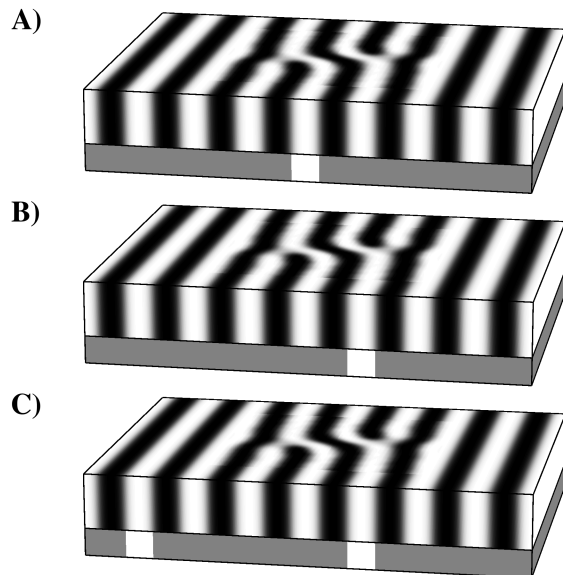


Figure 3.1: Illustrations of three different patterned underlayers used in this work. The top portion of each illustration is the BCP film with a dislocation defect of order 1. The bottom portion represents the brush underlayer. The background region is represented by the gray region and is neutral to both the white and black blocks of the BCP. The pinning stripe is the white region and is highly preferential to the white block of the BCP. A) An 8x density multiplying underlayer with pinning stripe position 0. B) An 8x density multiplying underlayer with pinning stripe position 2.

3.2.2 Thermodynamic Integration

Thermodynamic integration is a method that can be used to calculate the free energy difference between two different states in a molecular dynamics simulation. In thermodynamic integration the molecular dynamics simulation is brought from one state to another in a reversible manner. In this paper, thermodynamic integration is used to calculate the difference in the free energy of a defective BCP film versus a defect free lamellar state. The thermodynamic path used is summarized in Figure 3.2. A simulation is started in the mixed state at $\chi = 0$. This simulation is then phase separated using only an external potential by ramping the strength of the external potential from $A = 0$ to $A = 1$. Next χ is ramped from a value of $\chi = 0$ to $\chi = 0.55$. Finally, the external potential is turned off (by ramping $A = 1$ to $A = 0$). This is performed once using a defect free external potential and a second time

using a defect external potential.

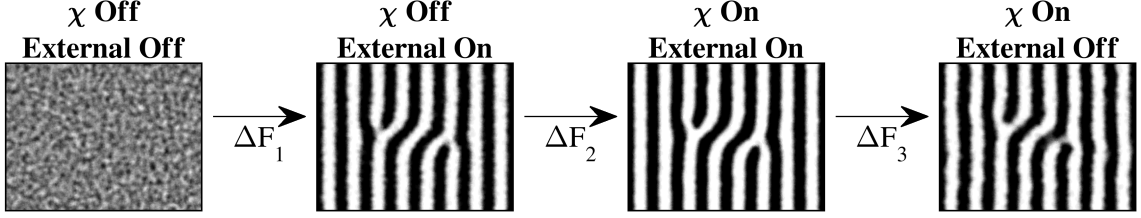


Figure 3.2: Images of the thermodynamic integration pathway used. Simulations start in a mixed state and then phase separated using an external potential. In the next branch χ is turned on which primarily just sharpens the interfaces. Finally the external potential is turned off which allows the film to relax and reach its equilibrium state.

The external potential used to generate defect free lamellae is given by the following equation:

$$V(x) = A \cdot \frac{\tanh \left[\frac{1}{2\pi w} \cos \left(\frac{2\pi}{L_0} \cdot x \right) \right]}{\tanh \left[\frac{1}{2\pi w} \right]}, \quad (3.2)$$

where V is the external potential, x is the x-position in the film, A is the magnitude of the external potential, w is related to the width of the interface (0.1 for this work), and L_0 is the pitch of the BCP. This potential can be seen in Figure 3.3 in the defect free case, as well as in the area outside the gray boxes in all the other potentials. The numbers at the bottom represent the relative position of each lamellae (normalized by $0.5 \cdot L_0$) to the center of the defect.

The external potential used to generate defective lamellae requires the use of a table potential, which can be seen inside the gray box in Figure 3.3. The tables for these defects were generated using a defect that formed naturally in a previously run simulation that started from a mixed state and was allowed to undergo phase separation on its own (i.e. without the aid of an external potential). The defect used naturally occurred and persisted for a long period of time. To generate the defect table, snapshots were taken of the simulation through time. These snapshots were then averaged through time in order to

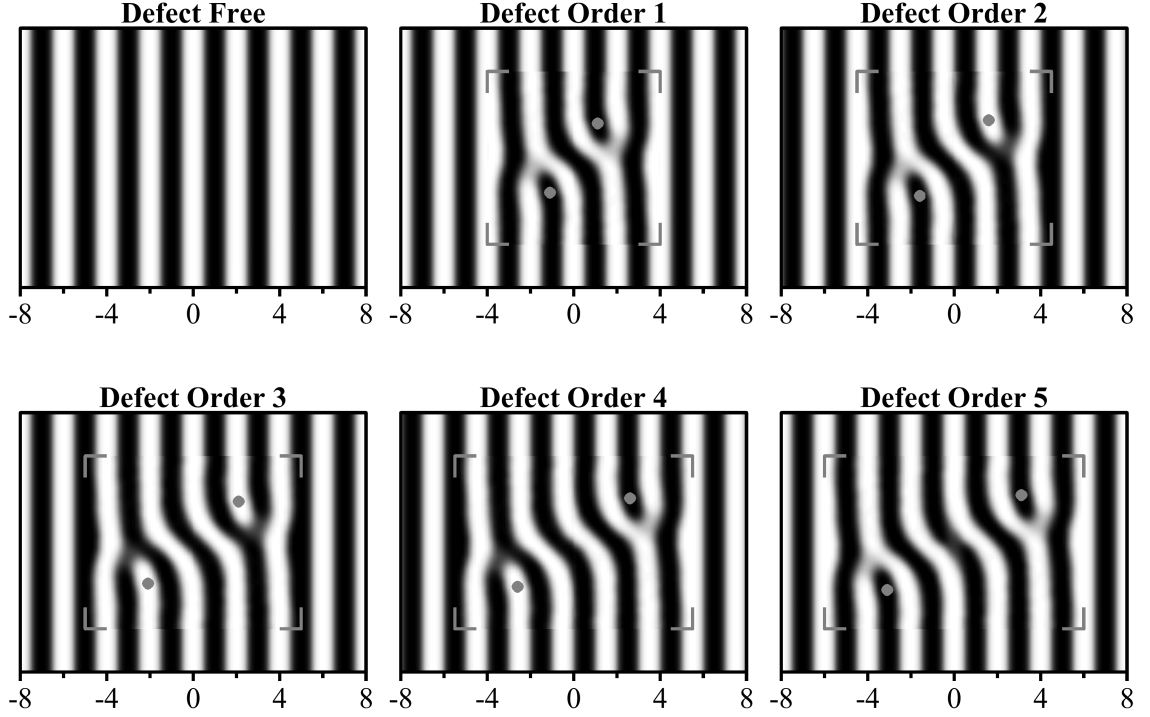


Figure 3.3: All external potentials used in this paper. The portion of the external potential outside the gray box is generated using Equation 3.2 while the area inside is generated using a table lookup that is interpolated with bicubic interpolation. The dislocation order indicates the number of lamellae between the two terminating blocks in the film, which are indicated by the gray dots. The x-axis indicates the position relative to the center of the defect, which is in units of number of lamellae.

generate the average defect state. It was then assumed that the volume fraction could be related to the potential directly. The tables used had a node spacing of $0.08 \cdot L_0$ with bicubic interpolation being used between nodes.

In the first branch, the external potential is turned on by ramping A from $A = 0$ to $A = 1$. It was found that $A = 1$ is sufficient strength for the external potential to fully phase separate the BCP film. The external potential was turned on in 50 steps, with a stepsize of $\Delta A = 0.02$. Each step in A was run for 1,000,000 timesteps, or 50 ns. It was found that equilibrium was typically reached within the first few nanoseconds. The external potential in the simulation was then logged every 100 timesteps. The external potential energy was averaged over the final 45 ns of each step. The integral in Equation 3.3 was then evaluated

using the trapezoidal rule.

$$\Delta F_1 = \int_{A=0}^{A=1} \frac{\langle V_{ext}(A) \rangle}{A} dA, \quad (3.3)$$

where ΔF_1 is the free energy difference between the beginning and ending state of the first branch and $\langle V_{ext}(A) \rangle$ is the average external potential measured when the external strength is A .

The second branch ramps on χ while keeping the external potential at full strength. The value of ε_{AB} is decreased in this branch in increments of $\Delta\varepsilon_{AB} = 0.005$ kcal/mol. Each step in ε_{AB} is run for 80,000 timesteps, or 4 ns. Every 100 timesteps the non-bonded potential between A and B beads is measured. This value is averaged over the final 2.5 ns to give $\langle V_{non-bonded}^{AB}(\varepsilon_{AB}) \rangle$, which is then used in the following integral to get the free energy of this branch:

$$\Delta F_2 = \int_{\varepsilon_{AB}=0.5}^{\varepsilon_{AB}=0.35} \frac{\langle V_{non-bonded}^{AB}(\varepsilon_{AB}) \rangle}{\varepsilon_{AB}} d\varepsilon_{AB}, \quad (3.4)$$

where ΔF_2 is the free energy difference over the second branch. This branch requires less timesteps at each step in ε_{AB} because little bulk rearrangement is happening in the film during this time. Instead, the primary changes noticed are a sharpening of the interfaces between lamellae.

The final branch ramps down the external potential strength. The value of A is decreased from $A = 1$ to $A = 0$ in 30 steps. Each step in A is run for 80,000 timesteps, or 4 ns. Every 100 timesteps the non-bonded potential between A and B beads is measured. This value is averaged over the final 2.5 ns to give $\langle V_{ext}(A) \rangle$, which is then used in the following integral to get the free energy of this branch:

$$\Delta F_3 = \int_{A=1}^{A=0} \frac{\langle V_{ext}(A) \rangle}{A} dA, \quad (3.5)$$

where ΔF_3 is the free energy difference over the third branch. Like the second branch,

less timesteps at each step is required because little bulk rearrangement is happening in the film. The change in free energy over this path, ΔF_i is then the sum of the parts, Equation 3.6. ΔF_i is the free energy of the defect or defect-free state after going through the path outlined. The relative free energy of the defect, ΔF , is then calculated by the difference of ΔF_{Defect} and $\Delta F_{Defect-Free}$.

$$\Delta F_i = \Delta F_1 + \Delta F_2 + \Delta F_3, \quad (3.6)$$

3.3 Results and Discussion

3.3.1 Pinning Stripe Location

Figure 3.4 shows the free energy difference for an 8x density multiplying underlayer for various pinning stripe positions. In these cases, there is only one pinning stripe in the system. The position of the pinning stripes is their distance from the center of the defect (divided by $0.5 \cdot L_0$), as was shown in Figure 3.3. It should be noted that odd numbered defects have pinning stripes centered on integer values (n), while even numbered defects have pinning stripes on the half values ($n + 0.5$). The individual branches of the thermodynamic integration for the second branch (ramping up χ) and third branch (turning off the external potential) branch are shown in Figure 3.4A and Figure 3.4B respectively, while the overall free energy difference is shown in Figure 3.4C. The points shown are the average of three replicates.

Looking at the overall free energy in Figure 3.4C it can be seen that the dislocations have similar trends with the exception of the dislocation with a defect order (DO) of 1. This is due to many DO = 1 simulations annealing out during the third branch, that is to say, the defects were annihilated leaving behind straight lamellae. Evidence of this can be seen in the third branch of the thermodynamic integration (Figure 3.4B). During the third branch, there should be almost no rearrangement of beads in the system if the defect that was built using the external potential is close to the true equilibrium defect shape. If there is no rear-

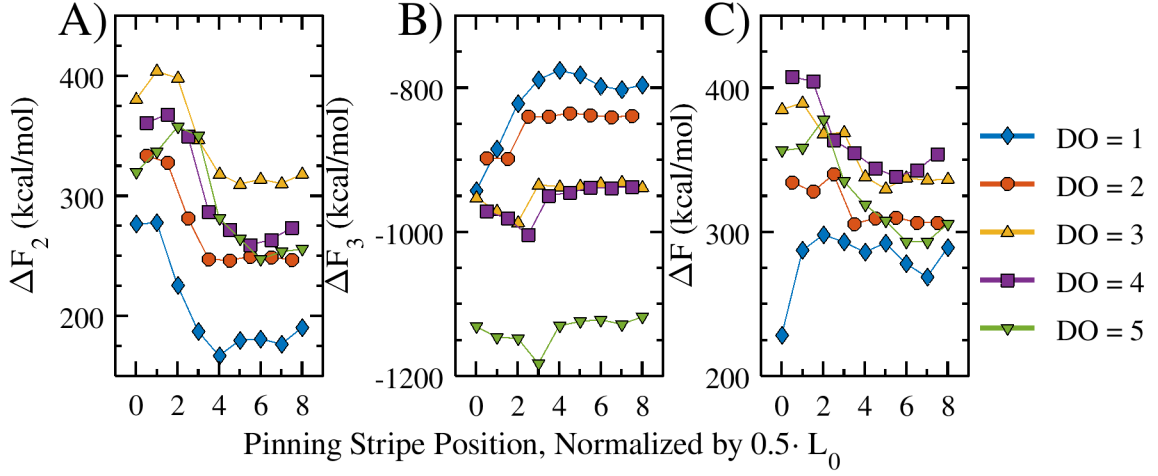


Figure 3.4: Plots showing A) the second branch of the free energy calculation, B) the third branch of the free energy calculation, and C) the total free energy difference between the defect and a defect free state versus the position of the pinning stripe for an 8x density multiplying underlayer.

rament, the third branch should be constant with respect to the pinning stripe position. This is approximately the case for all defects other than $DO = 1$, as will be discussed further later. When the pinning stripe approaches the center of $DO = 1$, the defect will frequently anneal out while ramping down the external potential. This means that the external potential is aiding in defect annihilation, increasing the kinetics for this to occur likely due to the small size of $DO = 1$. Longer time steps in the thermodynamic integration would likely make this effect worse and stands as one of the limitations of thermodynamic integration for looking at potentially unstable states. This spontaneous transition violates reversibility which invalidates the free energy measurement from thermodynamic integration in these cases.

On the other hand, for defects with $DO > 1$ the third branch is mostly constant with respect to pinning stripe position (PSP). There are some variations, in particular a lowering of ΔF_3 when the pinning stripe approaches the terminating block of the defect. In these cases the defect will necessarily adapt its shape slightly to accommodate the pinning stripe. However, these variations are small in magnitude and happen gradually in a reversible

manner, allowing thermodynamic integration to still be valid. Since the ΔF_3 is roughly constant with regard to pinning stripe position, and what few variations are present appear small, it can be assumed that the second branch of the integration, ΔF_2 , is the primary contributor to the total free energy with respect to the pinning stripe position. Therefore, while the total free energy of $DO = 1$ is likely invalid, the second branch of the integration can be looked at as a proxy for the total free energy.

A few observations can be made by analyzing the second branch of the integration (Figure 3.4A). First, as the pinning stripe moves further from the defect, towards higher PSP values, the free energy begins to level off at its minimum value. This is due to the pinning stripe interacting less and less with the defect, and therefore having a negligible effect on the free energy. The pinning stripe position where the free energy levels off increases with increasing defect order, but appears to be within $0.5 \cdot L_0$ of the terminating block. While $DO = 1$ levels off around $PSP = 3$, each increase in defect order seems to increase this transition by approximately 0.5 PSP. This suggests that pinning stripes beyond a distance of L_0 outside of the terminating block (marked by the gray dots in Figure 3.3) have little effect on the free energy of the defect.

As the pinning stripe approaches the defect, the free energy increases greatly due to the increased interactions of the pinning stripe with the defect. The magnitude of this increase is on the order of 50 to 100 kcal/mol, implying that these defects will almost never occur naturally above the pinning stripe at equilibrium.

For some of the larger defects, such as $DO = 3$ or 5, there appears to be a maximum in the free energy when the pinning stripe is not at the center of the defect ($PSP = 0$). The position of these maximums are roughly $PSP = 1$ and 2 for $DO = 3$ and 5 respectively, implying the maximum likely shifts further right as defect order is increased. The location of the maximum corresponds to the lamellae interior to the terminating block of the defect, which is at $PSP = 2$ and 3 for $DO = 3$ and 5 respectively. Since the pinning stripe being directly under or to the interior of the terminating block consistently gives the maximum

in free energy, it suggests a pinning stripe in these locations destabilizes the defect. This pinning stripe location also likely has the highest kinetics for defect annihilation since a pinning stripe in this location will increase the likelihood of bridge formation which is a necessary step in defect annihilation.[90]

The location where the free energy levels off can be a good approximation for what density multiplication is large enough to allow the defect to form. For example, since $DO = 1$ levels off around $PSP = 3$, it can be assumed that this dislocation is more likely to form if the density multiplication is 3x or greater since the defect can form entirely between two pinning stripes. However, if the density multiplication is less than 3x then there is guaranteed to be a pinning stripe in either $PSP = 0, 1$ or 2 , which are in the increased free energy region. Similarly, for $DO = 3$ the free energy levels off around $PSP = 4$ and for $DO = 5$ it levels off around $PSP = 5$. This implies that decreasing the density multiplication should decrease the number of the larger defects significantly. However, since the maximum in the free energy is not at $PSP = 0$ for these larger defects, there exists the possibility of a dislocation pair that straddles a pinning stripe. For example, with $DO = 5$, if the pinning stripe is at $PSP = 0$, the dislocation pair can fit nicely on a 3x underlayer with one pinning stripe in the center of the defect and another two on the outside of the dislocation pair (at positions -6 and $+6$). This particular example defect has been observed naturally in simulations using this model and has been shown to be very stable since the defect persisted without changing in defect order for very long simulation times.

Figure 3.5 looks at the overall free energy for unpatterned underlayers (infinite density multiplication). It can be seen that the free energy is lowest for $DO = 1$, increases for $DO = 2$, increases further for $DO = 3$, stays approximately the same for $DO = 4$, and then decreases for $DO = 5$. It is hypothesized that this trend is due to two competing factors. First, increasing the defect order increases the amount of curvature in the BCP system which enthalpically would increase the free energy. Second, the two halves of the dislocation pair likely destabilize each other when they are too close. This hypothesis

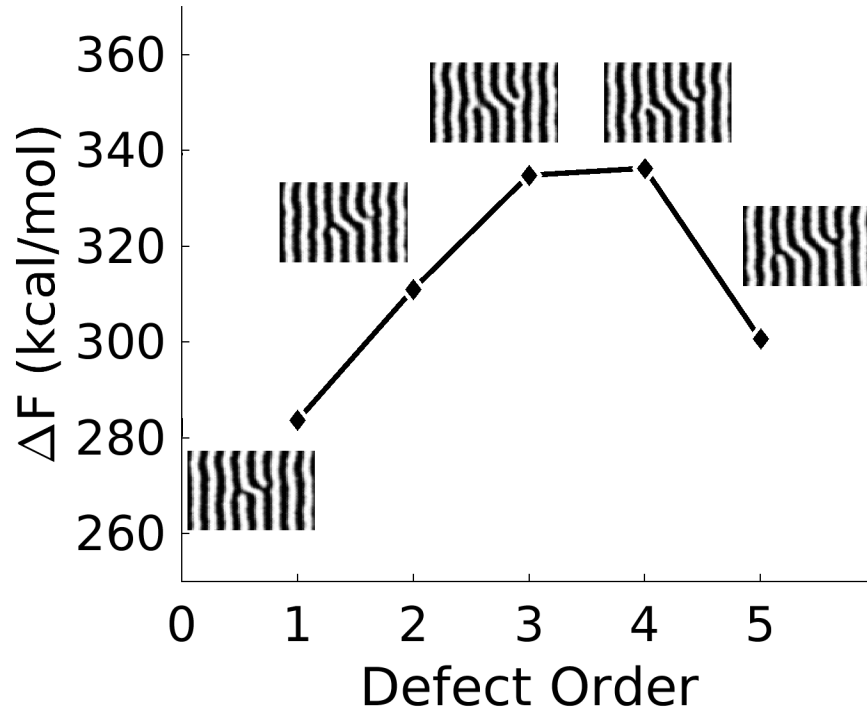


Figure 3.5: Plot showing the effect of defect order with the free energy difference between a defect and defect-free state.

would lead to the observed results where initially the free energy increases from defect free to DO = 1 to DO = 3 due to the increasing interfacial area. When DO = 4 is reached, the increase in interfacial area is counteracted by the stabilizing effect of the dislocation pairs being further separated. Finally, with DO = 5 the defect begins to get more stable due to the further separation of the dislocation pairs. This has large practical consequences on defect annihilation kinetics. If it is assumed that the pathway to annealing DO = 5 is for the defect to first shrink to DO = 4, then to DO = 3, and so on, as has been observed in simulation,[90, 109] then DO = 4 could be considered a transition state in the process of annealing DO = 5 to a defect free state. While DO = 5 has a much higher free energy than defect free (≈ 300 kcal/mol) and should therefore be virtually non-existent at equilibrium, the fact that the transition state DO = 4 has a far higher free energy (≈ 340 kcal/mol) suggests there will be very slow kinetics in annealing out DO = 5. This data helps support the theory that defects

in BCP films are kinetically trapped.

It has been shown that thermodynamic integration simulations can suffer from a limited simulation volume.[92] This was a concern, particularly for the larger defect orders since it is more likely the defect could see itself across the periodic boundary. Though it should be noted that the presence of a pinning stripe near the periodic boundary of the system lessens this concern since the pinning stripe decreases the amount a defect could influence itself across the boundary. In order to test the simulation volume effect, a limited number of larger simulations were run. These larger simulations were $12 \cdot L_0$ wide, while the typical simulations were only $8 \cdot L_0$ wide. A single replicate was run for defect orders 3, 4, and 5 where the size effect should be most apparent. This is also where the free energy begins to level off and then decrease as detailed in the previous paragraph, so these larger simulations should help verify that trend. An underlayer with 12x density multiplication was used, with the pinning stripe located away from the defect on the edge of the simulation. The results of the larger volume simulations showed the same general trends as was observed in the smaller simulations, with $DO = 3$ and $DO = 4$ having roughly equivalent free energies ($\Delta F = 256$ and $\Delta F = 250$ kcal/mol respectively) and $DO = 5$ having significantly lower free energy ($\Delta F = 170$ kcal/mol). However, while the trend is the same the absolute values of the free energy differences were approximately 100 kcal/mol lower than before. This suggests that the smaller simulations are indeed experiencing this size effect, however it appears they do capture trends accurately.

3.3.2 Density Multiplication

Underlayers with 1x, 2x, 4x, and 8x density multiplication were simulated. Additionally an unpatterned underlayer was simulated in order to approximate an infinite density multiplication. These results are shown in Figure 3.6 where the free energy differences between a defective film and a defect free film is shown for various defect orders at as a function of density multiplications. The points shown for each density multiplication are the free

energy difference corresponding to the lowest average free energy over all pinning stripe positions for that density multiplication. It can be seen that as the density multiplication increases the difference in free energy decreases. This is due to higher density multiplications having less pinning stripes per area and therefore a lowered driving force for pattern correction when a defect is present. Intuition would suggest that the free energy difference should asymptotically approach the unpatterned (infinite density multiplication) case as the density multiplication increases, which appears to be the case in these simulations.

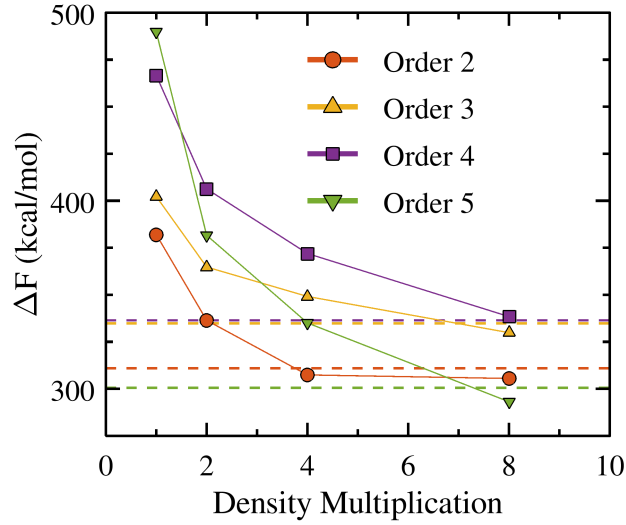


Figure 3.6: Free energy differences for thermodynamic integration for various density multiplications. Each point represents the minimum free energy difference of all pinning stripe positions for that density multiplication. The horizontal dashed lines near the bottom represent the free energy difference of the defect measured on an unpatterned underlayer.

Assuming a Boltzmann's distribution, the defect density can be estimated using the free energy difference and Equation 3.1. From Figure 3.6 these free energy differences can be used to determine what the return is for putting in the increased lithographic effort to make a more highly defined underlayer pattern. For instance, for $DO = 5$, the difference in free energy between 1x and 2x density multiplication is approximately 100 kcal/mol. Since in these simulations $k_B \cdot T \approx 1$ kcal/mol, it is e^{100} times more likely to find this defect on a 2x density multiplication than 1x. This type of comparison can be useful when looking at higher density multiplications to determine when increasing density multiplication will

no longer have an effect on the equilibrium defect density. For all defects shown here the 8x underlayers had nearly the same defect free energy difference as the unpatterned cases, meaning that at equilibrium the defect density for any density multiplication greater than 8x will all be nearly the same. With the exception of DO = 2, decreasing the density multiplication from 8x to 4x increased the free energy difference by 20 kcal/mol or more. This suggests these defects will occur at least $5e8$ times more on an 8x underlayer than a 4x underlayer. On the other hand, for DO = 2, the defects are still small enough that a decrease in density multiplication from 8x to 4x has no effect on the free energy, and even higher smaller density multiplications would be needed to affect the free energy.

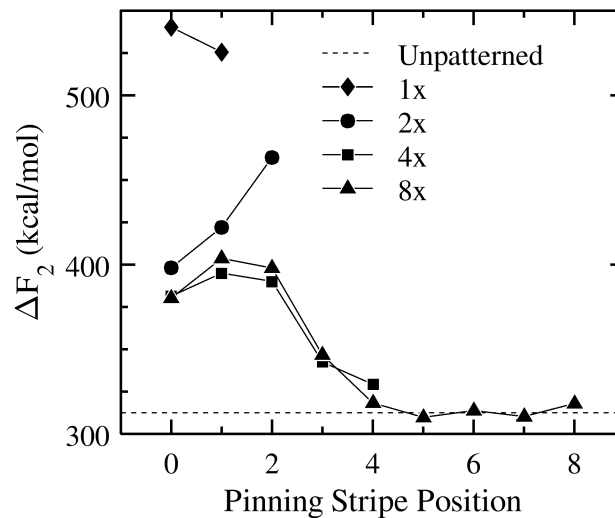


Figure 3.7: Free energy differences for the second branch in the thermodynamic integration for DO=3. The unpatterned underlayer free energy is represented by a horizontal dashed line at the bottom of the graph.

Figure 3.7 takes a closer look at the effect of density multiplication in regards to pinning stripe position for DO = 3 by looking at the second branch of thermodynamic integration. The previous section has already discussed the 8x underlayers and have explained why having a pinning stripe in positions 1 or 2 (PSP = 1 or PSP = 2) has the highest free energy difference due to its proximity to the terminating block. As the density multiplication decreases the number of pinning stripes present in the underlayer increases. For 4x un-

derlayers there are two pinning stripes throughout our simulation, yet the 4x underlayers mostly follow the same trend as 8x underlayers. This implies that the extra pinning stripe is still too far away from the defect's terminating blocks to cause it any substantial instability.

For 2x underlayers four pinning stripes are present in the simulations underlayer. It can be seen that the free energy significantly increases as the PSP shifts to the right. This trend can be explained by looking at the locations of all the pinning stripes in the system, but especially the two closest to the defects terminating blocks. For 2x density multiplication, the pair of pinning stripes are always $PSP = x$ and $PSP = x \pm 4$ (See Figure 3.3, Defect Order 3 for reference). In the case of $PSP = 0$ and ± 4 , the closest distance from one pinning stripe to a terminating block is relatively far (L_0). On the other hand, $PSP = 1$ and -3 has both pinning stripes $0.5 \cdot L_0$ away from the terminating block (one on the inside and one on the outside). While in the 8x simulations a pinning stripe in position 3 does not significantly increase the free energy, a pinning stripe in position 1 does, explaining why this point is higher in free energy than $PSP = 0$ and ± 4 . A similar analysis can be done for $PSP = 2$ and -2 . Since both of these pinning stripes are beneath a terminating block, and from the 8x simulations we see that this is near where the maximum in the free energy occurs, it follows that this underlayer would have the highest free energy of the 2x underlayers.

A similar analysis can be performed for the 1x underlayer. In this case the nearest pinning stripes are at $PSP = x \pm 2$. Using this, when $PSP = 0$ and ± 2 there are two pinning stripes directly underneath the terminating block as well as one pinning stripe in the center of the defect which contributes greatly to increasing the free energy. When $PSP = -3, -1, 1,$ and 3 while there are two pinning stripes in highly unfavorable spots ($PSP = \pm 1$) the two other pinning stripes are getting to the region where they will have less effect on the free energy, causing this position to have an overall lower free energy.

3.4 Conclusions

Molecular dynamics simulations were implemented to determine the effect the pinning stripe and density multiplication has on the free energy of dislocation defects of various orders. For all defect orders it was found that the free energy was generally highest when the terminating block of a dislocation was situated above a pinning stripe, suggesting there is little probability of the defect being positioned in these locations at equilibrium. It was found that pinning stripes approximately $1 \cdot L_0$ outside of the terminating blocks of the defect have little effect on the free energy of the defect. If the defect is large enough, there exists a location in the center of the defect where pinning stripes have less of a free energy penalty than underneath the terminating block, suggesting defects can be stable while straddling a pinning stripe. For the defects explored here when the density multiplication was 8x, the thermodynamic driving force for the removal of the defects simulated here was approximately the same as for unpatterned underlayers because the defect was able to fit in an area without a pinning stripe nearby. While increasing density multiplication does theoretically increase the number of defects present due to a decreased free energy difference, the free energy difference is still incredibly high even on an unpatterned underlayer, making their estimated population very small. This bolsters the idea that the high defect densities reported experimentally are due to kinetic entrapment. Increasing the defect order does not appear to increase the free energy monotonically. Instead, a defect of order 5 appears to have a lower free energy than a defect of order 3. It is hypothesized that free energy versus defect order is controlled by two competing factors: the increase in interfacial area for larger defects increases free energy while the greater separation of the dislocation pairs decreases free energy by stabilizing each dislocation.

CHAPTER 4

EFFECT OF HOMOPOLYMER ON LINE EDGE- AND LINE WIDTH-ROUGHNESS FOR BCP/HOMOPOLYMER BLENDS

4.1 Introduction

In the previous chapter a relative defect free energy for various dislocation defects was calculated using molecular dynamics simulations. Looking top-down on a phase separated BCP film, it's relatively easy to point out defects such as dislocations and determine the utility of the BCP film for lithographic application. However, being defect-free is not the only qualifier that a phase separated BCP film needs to be ready for implementation into the chip manufacturing process. Two additional factors that are used to gauge the utility of DSA of BCPs are the lamellae's line edge roughness (LER) and line width roughness (LWR). LER is the variation in the position of the lamellae's interface, while LWR is the variation in the width of the lamellae. High LER and LWR lamellae are known to negatively affect the performance of electronics patterned by them.[94, 95, 110] For instance, if a gate (a potential feature to be patterned by BCPs) between a source and drain were to be patterned with a high LER and LWR it could affect the transport of electrons through the gate (e.g. current leakage). Unlike defects such as dislocations which can be spotted from an SEM image, LER and LWR require good quality images of the lamellae to go through an image analysis to determine their values, making them potentially more subtle characteristics of the lamellae than defects.

Simulations have shown that for chemoeitaxial DSA of BCPs, factors known to affect the BCP LER include background composition (f_A) and pinning stripe width.[78] However these simulations were done with a pure BCP thin film. Homopolymer is a known additive for BCP powders that, depending on the ratio of BCP/homopolymer, can tune the

achievable pitch of the BCP. In this chapter molecular dynamics simulations will be used to show the affect that various amounts of homopolymer has on LER and LWR for lamellae forming BCPs. For an A-B block copolymer, homopolymer of the A-block and B-block will be added in equal amounts to prevent a change in the effective volume fraction of the BCP. Dynamics are run on pre-aligned lamellae thin films with various pinning stripe widths, background compositions, density multiplications, and homopolymer concentrations (n_{Homo}). LER and LWR are then measured using in the thin films at the top 5 nm and bottom 5 nm and reported.

4.2 Experimental Methods

The work in this chapter employs the model described in Chapter 2 for molecular dynamics simulations. All BCPs in this work were an $N = 64$ (16-bead chains) with a 50/50 ratio of A/B beads (8 A-beads, 8 B-beads). To generate homopolymer in an A-B BCP film, first an A-B film is generated. Next, chains are broken apart into their constitutive homopolymers until the desired homopolymer loading is present. To break apart a chain, the bond between the 8th and 9th beads must be removed, as well as the angle potential between 7-8-9 and 8-9-10. Homopolymer chains have the same properties and parameters as their A-B counterparts including the same ϵ_{ij} . This means that the homopolymer is $0.5 \cdot N$ of the BCP chains. The homopolymer concentration n_{Homo} is defined here as the percentage of chains that are homopolymer.

All films used in this study were pre-aligned before dynamics were run. To achieve this, BCP chains are rotated such that the primary axis of the radius of gyration tensor is placed along the x dimension, the secondary axis is placed along the y dimension, and the tertiary axis is placed along the z dimension. The chain's center of mass is then placed along where a lamellae interface is expected to be.

Since homopolymer addition causes an increase in the BCP's pitch, the nominal pitch of the BCP/homopolymer blend needs to be measured. To do this, pre-aligned bulk NPT

simulations of BCP/homopolymer blends were generated with ten lamellae. The bulk simulation had an x dimension (the dimension perpendicular to the lamellae's direction) of $5 \cdot L_{est}$ where L_{est} is an over or under estimate for the pitch of the BCP/homopolymer blend. In the NPT simulation the volume is not held constant, meaning each dimension can vary in size to accomodate the BCP/homopolymer blend. The y and z dimensions are coupled, meaning they will change by equal amounts, while the x dimension was allowed to change independently of the other two. The simulation was allowed to run for 10 ns for various n_{Homo} over six replicates. An example of the changing x - and y/z - dimension is shown in Figure 4.1, showing that after 5 ns the simulation's x dimension is relatively stable. The x -dimension was averaged over the last 5 ns and the nominal pitch (L_o) for a particular BCP/homopolymer blend was determined by dividing this value by five (the number of lamellae pairs). Homopolymer concentrations used for this study were $n_{Homo} = 0, 0.05, 0.10, 0.18, 0.26$, and 0.33 . Figure 4.2 shows the relationship between n_{Homo} and L_o for this work.

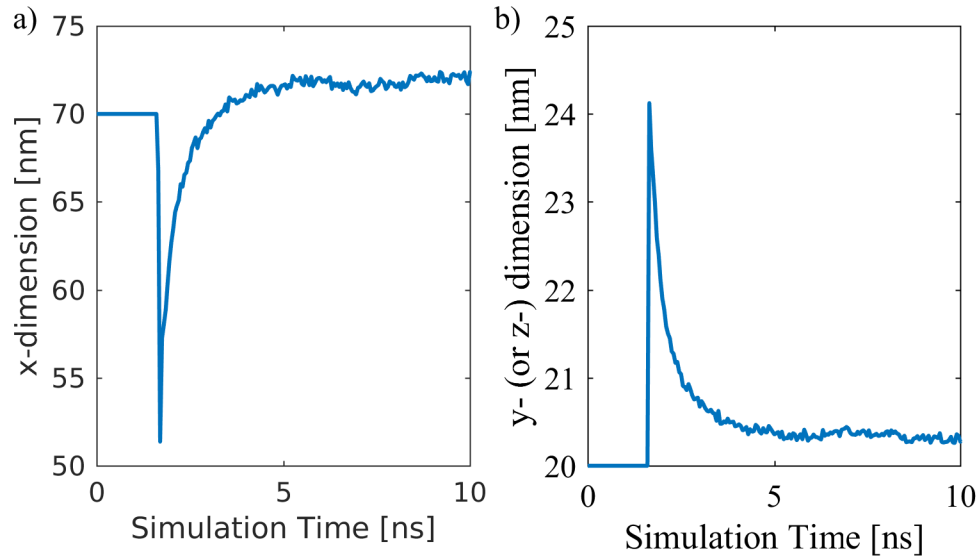


Figure 4.1: Example of an NPT simulation running to determine pitch. The x dimension (a) is allowed to vary uncoupled from the other dimensions to accommodate the ten pre-aligned lamellae. The y and z dimensions (b) were the same value initially and due to being coupled, vary equally with one another.

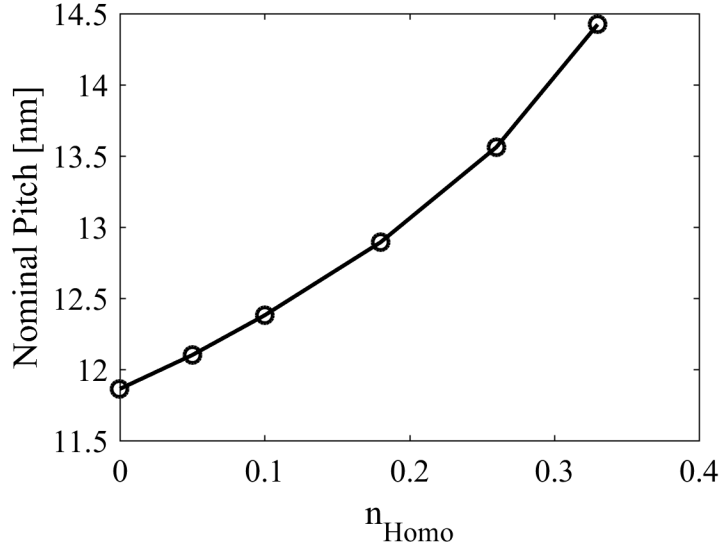


Figure 4.2: Plot showing how the nominal pitch of the blend increases as the fraction of homopolymer chains ($N = 8$) increases.

Thin film BCP/homopolymer simulations were run under NVT conditions on patterned substrates with either a 2x or 3x multiplication density. The simulation size for each homopolymer variation was $6L_o \times 100 \text{ nm} \times 1L_o$. Background compositions (f_A) explored were 0.30, 0.40, and 0.50 while pinning stripe widths used here were $0.3L_o$, $0.4L_o$, $0.5L_o$, $0.6L_o$, and $0.7L_o$. Underlayers are patterned by selectively changing the bead types in the underlayer. Pre-aligned simulations were allowed to run for 80 ns. LER and LWR were calculated as described in Chapter 2 and averaged over the last 20 ns of each simulation. Six replicates were run for each simulation condition and the means are reported. Confidence intervals (95%) for LWR and LER ranged from 0.02 - 0.12 for $n_{Homo} = 0$ to 0.26. At $n_{Homo} = 0.33$, LWR and LER 95% confidence intervals were closer to 0.20.

4.3 Results and Discussions

Experimentally, SEMs take images of films but only probe through a specific depth at the surface. This means that the contrast seen in the SEM is really a representation of the average block concentration averaged over some surface thickness, rather than the entire

film. This is problematic because the LER and LWR are expected to change near the substrate where the patterned underlayer has more influence over the lamellae's structure. For these reasons the LER and LWR measurements were done for the top and bottom 5 nm of each BCP/homopolymer film. A distinction is also made between the LER and LWR of pinned and unpinned lamellae interfaces, given that the pinned lamellae will have higher levels of guidance by the preferential pinning stripe. A pinned lamellae interface is defined as one of the interfaces belonging to the pinned lamellae and the unpinned lamellae interfaces are the remaining.

4.3.1 Line Edge Roughness

Figure 4.3 shows the effect of n_{Homo} on the LER of pinned and unpinned lamellae interfaces at the top and bottom of the film for an $f_A = 0.30$. LER increases with n_{Homo} at the top and bottom of the film for both lamellae types. Little dependence is seen in LER with pinning stripe width with the exception of the unpinned lamellae interfaces near the substrate (Figure 4.3c) at $n_{Homo} > 0.10$. At the surface of the film unpinned (Figure 4.3a) and pinned (Figure 4.3b) lamellae interfaces show little difference between their measured LER. This is expected since the lamellae are only expected to behave differently due to the patterned substrate, but near the film surface, the lamellae are likely outside the influence of the substrate. However near the substrate, LER is shown to be higher for the unpinned interfaces (Figure 4.3c) compared to the pinned interfaces (Figure 4.3d) due to pinning stripe increasing the guidance of the pinned lamellae. The unpinned lamellae interfaces near the substrate shows larger variation in LER with n_{Homo} compared to any other observed lamellae LER. These unpinned lamellae interfaces at the substrate likely have more sensitivity to n_{Homo} due to larger homopolymer segregation occurring as n_{Homo} increases which can lead to larger variations in the placement of a lamellae interface. A deeper discussion on this will occur in a later section. Overall for low n_{Homo} deviations in the LER between the top and bottom of the film, for pinned and unpinned lamellae are minimal.

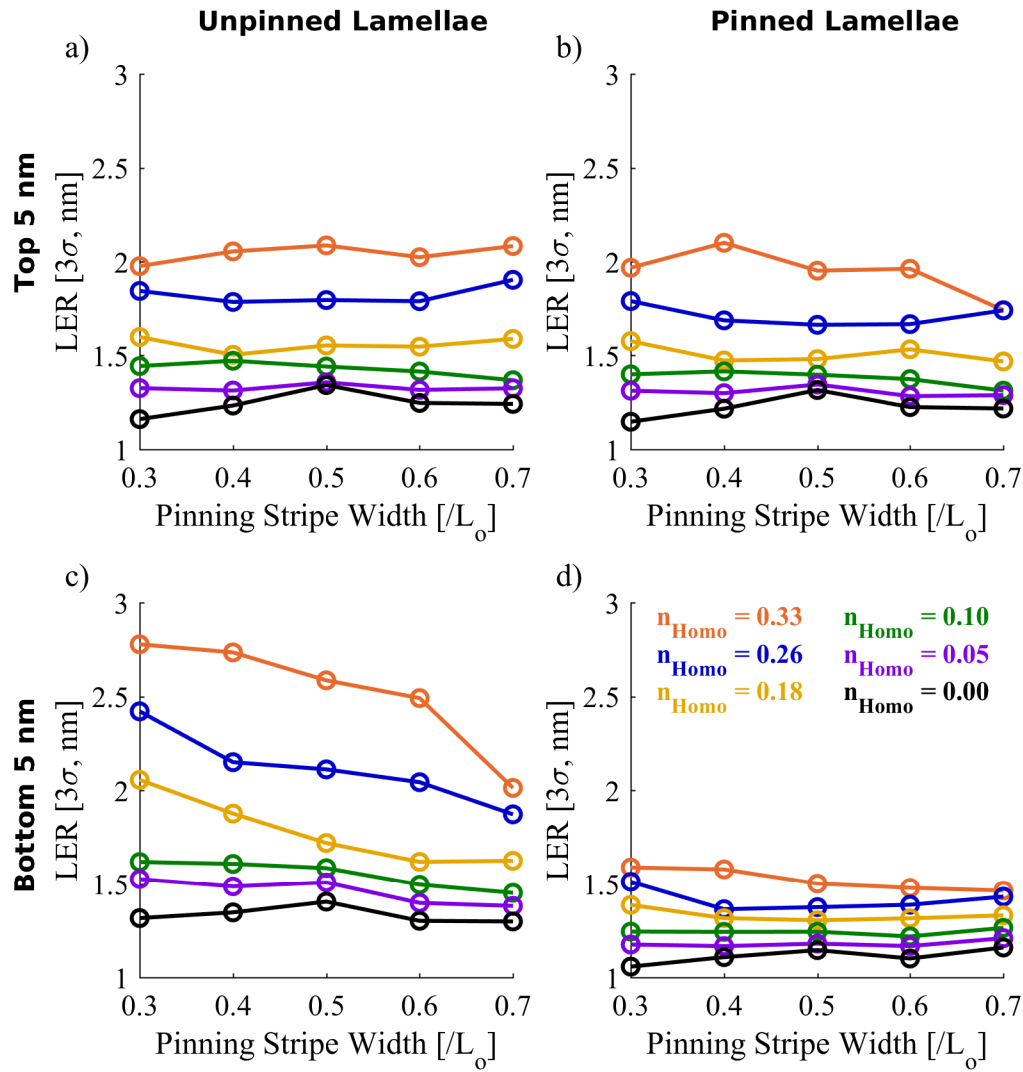


Figure 4.3: LER for the top (a & b) and bottom (c & d) 5 nm of the BCP/homopolymer film simulations for the unpinned (a & c) and pinned (b & d) lamellae for varying n_{Homo} . All data shown is for 2x density multiplication underlayers with an $f_A = 0.3$.

While simulations were run for $f_A = 0.30, 0.40$ and 0.50 , Figure 4.4 will display only $f_A = 0.30$ and 0.50 . This is done for ease of the viewer and as a representation of the near expected optimal background composition ($f_A = 0.30$) for a 2x density multiplication underlayer versus a neutral background composition ($f_A = 0.50$). An estimation for the optimal background composition (in terms of ensuring lamellae do not flip orientation) can be determined by the ratio of different lamellae over the region. For instance, for 2x underlayers, one A-block lamellae and two B-block lamellae are present over the background region, making it's optimal composition $f_A = 0.33$. Near the top of film, the unpinned (Figure 4.4a) and pinned (Figure 4.4b) lamellae interface's LER are relatively unaffected by the shift in f_A , showing nearly the same LER between the two different types of interfaces. This is due to the same reason as before, the lamellae interfaces here are far from the influence of the underlayer. Near the substrate pinned lamellae interfaces (Figure 4.4d) show an increase in LER as f_A approaches 0.50 . As f_A approaches 0.50 , the energetic contrast between the pinning stripe and the background region decreases, lowering the level of guidance for the lamellae near the substrate.[78] The unpinned lamellae do not show a significant change in LER at low n_{Homo} with respect to f_A since they are only in contact with the background region. At high n_{Homo} however a neutral background composition lowers the LER of unpinned lamellae near the substrate.

4.3.2 Line Width Roughness

If the placement of the lamellae interface varies along the lamellae, but each lamellae interface varies in the same manner, the lamellae interfaces are said to be correlated which leads to a high LER and a low LWR. An example of this is if each lamellae had the shape of a sine wave. However, if each lamellae instead had a peristaltic shape, the lamellae interfaces are said to be highly uncorrelated and this would lead to a high LER and LWR. This means that the LWR is more so a measure of how correlated the lamellae interfaces are to one another.

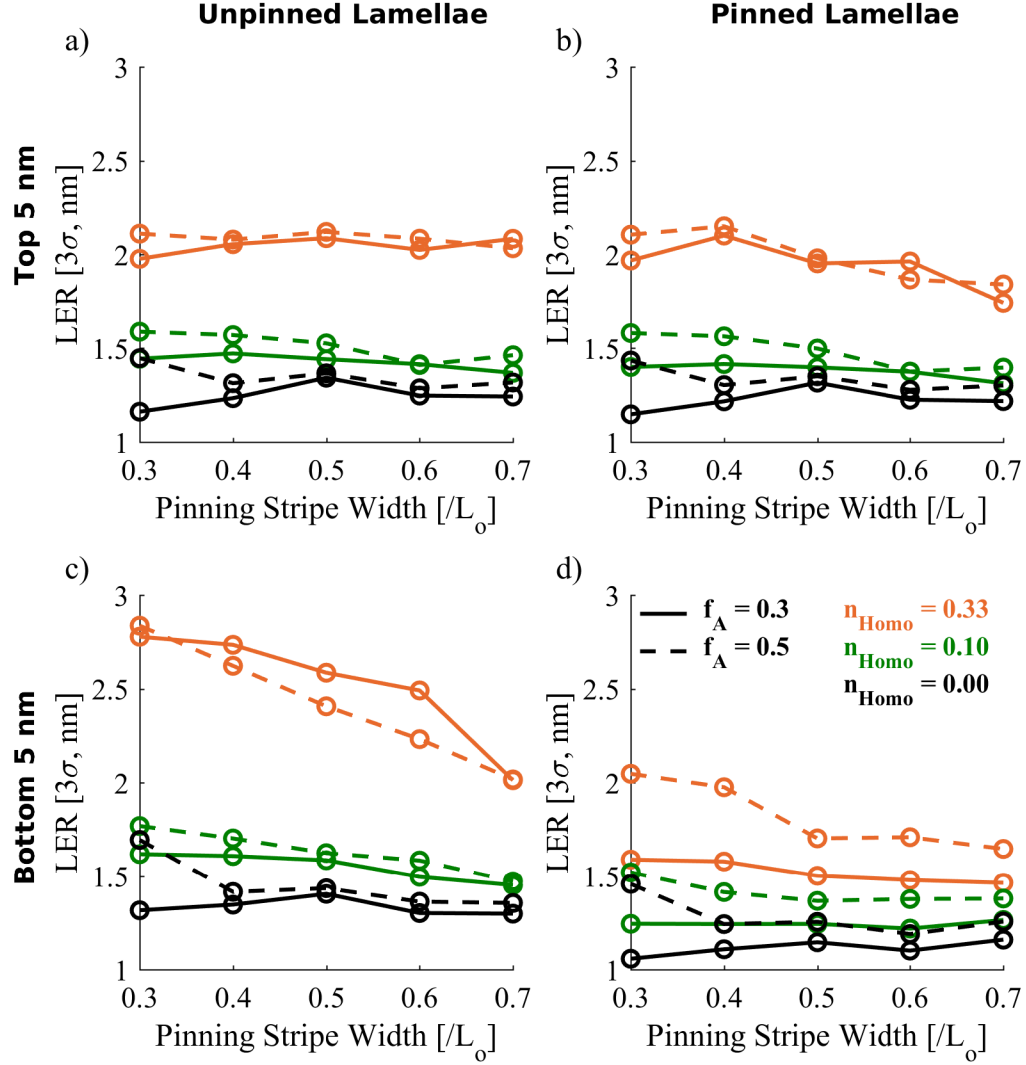


Figure 4.4: LER for the top (a & b) and bottom (c & d) 5 nm of the BCP/homopolymer film simulations for the unpinned (a & c) and pinned (b & d) lamellae for varying n_{Homo} at $f_A = 0.30$ (solid lines) and $f_A = 0.50$ (dashed lines). All data shown is for 2x density multiplication underlayers.

Similarly to LER, LWR increases in the BCP/homopolymer blends for all $n_{Homo} > 0$ at an $f_A = 0.30$ for 2x density multiplication underlayers (Figure 4.5). At the top of the film unpinned (Figure 4.5a) and pinned (Figure 4.5b) lamellae interfaces are fairly correlated with their adjacent interfaces at $n_{Homo} < 0.10$. However for $n_{Homo} > 0.10$ the unpinned lamellae interfaces become more uncorrelated with one another compared to the pinned lamellae interfaces. Near the patterned substrate, n_{Homo} has a similar influence on the LWR of the pinned (Figure 4.5d) and unpinned (Figure 4.5c) lamellae. Ultimately for low n_{Homo} the LWR is nearly equal between unpinned and pinned lamellae at both the top and bottom of the film, differing significantly between one another for only $n_{Homo} = 0.26$ and 0.33 . As n_{Homo} increases a trend can be observed with changing pinning stripe widths. This effect may be attributed to homopolymer segregation. The trend of increasing LWR with n_{Homo} may be explained by fluctuations in homopolymer concentrations along the lamellae. Both of these will be explained in the next section.

Figure 4.6 shows how the LWR is affected by changing f_A for 2x density multiplication underlayers. Background composition does not appear to affect the LWR at the top of the film for pinned (Figure 4.6b) and unpinned (Figure 4.6a) lamellae for low n_{Homo} . At high n_{Homo} near the top of the film as f_A becomes more neutral, LWR decreases. This too may be the influence of homopolymer segregation between the top and bottom of the film. Near the substrate, the unpinned lamellae (Figure 4.6c) LWR show little sensitivity for changing f_A , but at high n_{Homo} a more neutral f_A leads to a large increase in LWR. Similarly to LER, the pinned lamellae near the substrate (Figure 4.6) increases its LWR as the f_A approaches 0.50 due to a decrease in the energetic contrast between the pinning stripe and the background region.

4.3.3 Homopolymer

Homopolymer in blends tends to segregate to either the free surface or the substrate rather than forming a uniform distribution through the BCP (Figure 4.7). Due to the homopoly-

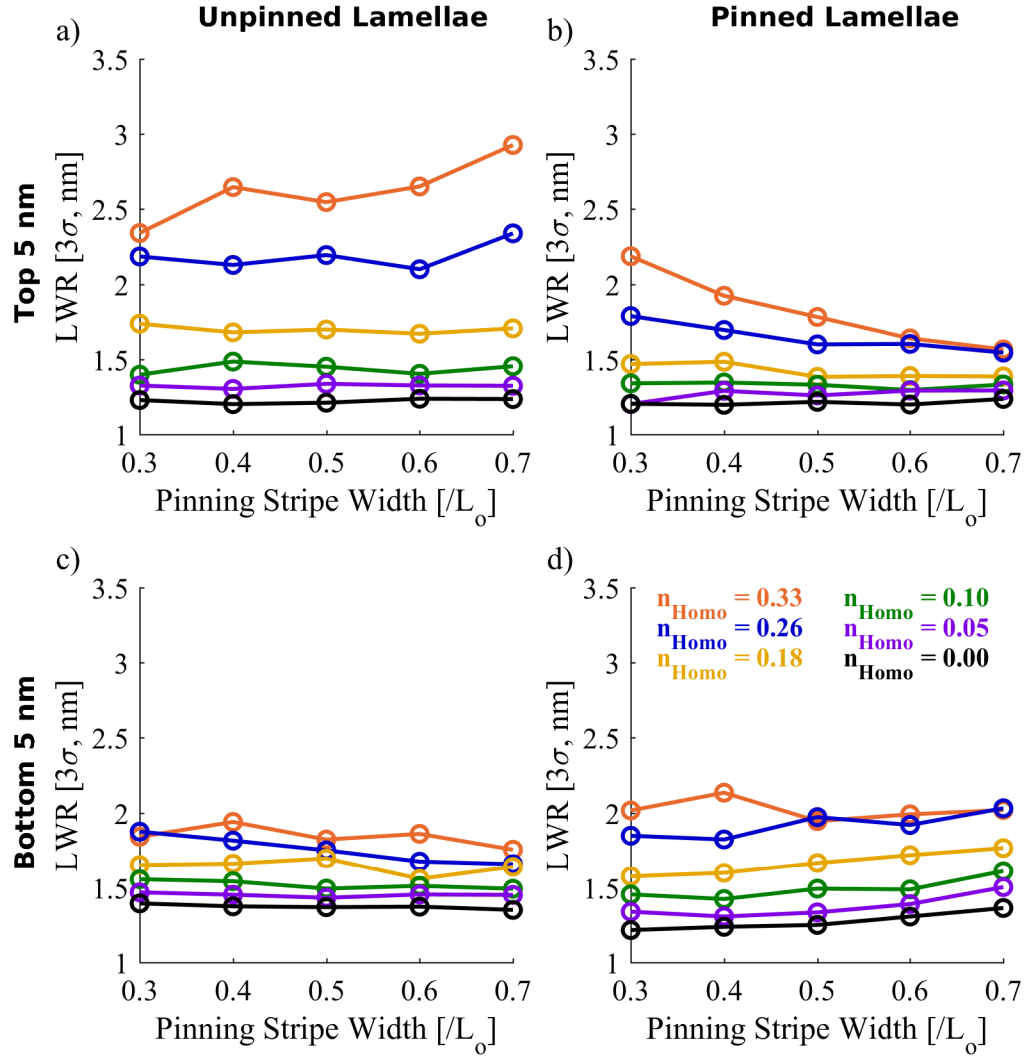


Figure 4.5: LWR for the top (a & b) and bottom (c & d) 5 nm of the BCP/homopolymer film simulations for the unpinned (a & c) and pinned (b & d) lamellae for varying n_{Homo} at $f_A = 0.30$. All data shown is for 2x density multiplication underlayers.

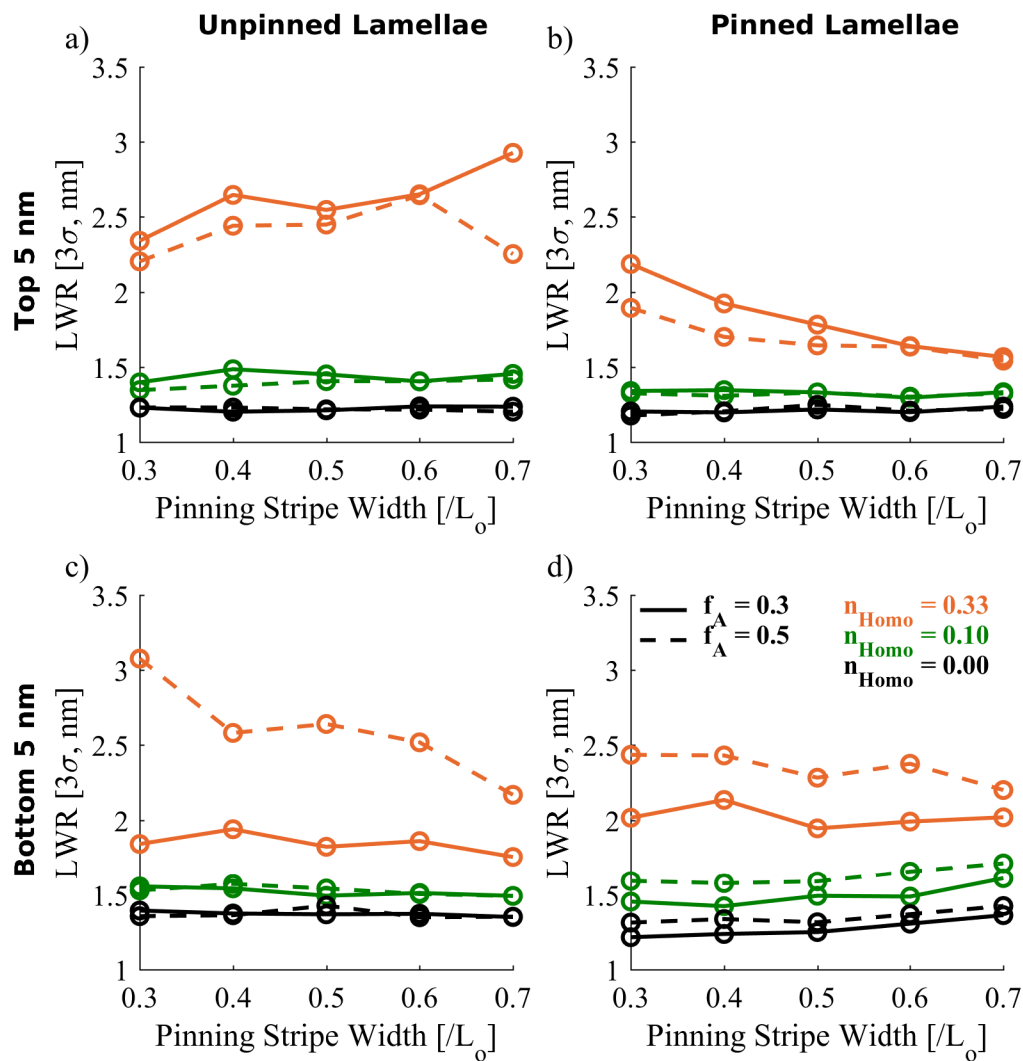


Figure 4.6: LWR for the top (a & b) and bottom (c & d) 5 nm of the BCP/homopolymer film simulations for the unpinned (a & c) and pinned (b & d) lamellae for varying n_{Homo} at $f_A = 0.30$ (solid lines) and $f_A = 0.50$ (dashed lines). All data shown is for 2x density multiplication underlayers.

mers' lower molecular weight ($0.5 \cdot N_{BCP}$) it has a preference to interact with the free surface. However, the homopolymer has a stronger preference to interact with underlayer, especially regions as preferential as the pinning stripes. For instance in Figure 4.7 it can be seen that for pinned lamellae (A-Block, white) over the pinning stripe a large portion of their homopolymer (lime green) is located near the bottom of the BCP film. For f_A 0.3 (Figure 4.7a) the unpinned A-Block lamellae homopolymer segregates to the free surface instead. The B-block unpinned lamellae (turquoise) has it's homopolymer (magenta) more evenly distributed at the top and bottom of the films and through the lamellae. This is because while the homopolymer does prefer the free surface, the background region is slightly preferential to the B-block leading to a more uniform distribution through the thickness of the lamellae.

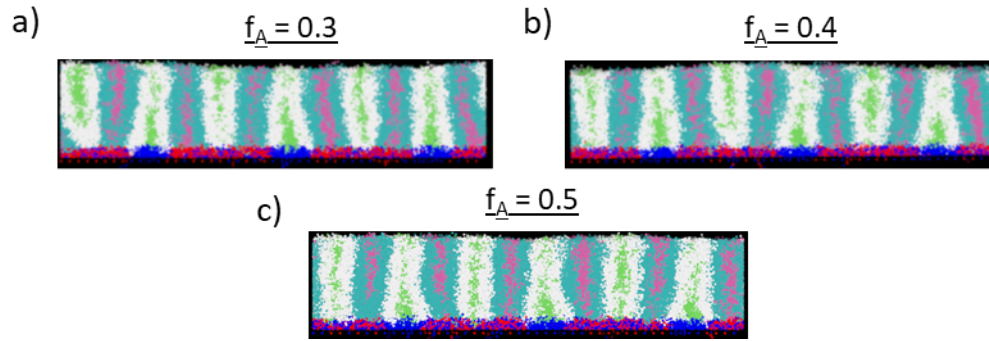


Figure 4.7: Cross sectional (xz plane) view of BCP/homopolymer blend film in simulation for $f_A = 0.30$ (a), $f_A = 0.40$ (b), and $f_A = 0.50$ (c). The turquoise and white beads are the BCP beads, while the magenta and lime green beads are their corresponding homopolymer.

As f_A approaches 0.50 in Figure 4.7, the unpinned A-block lamellae begins to form a more uniform distribution throughout its lamellae thickness. This is at the cost though of an undercutting of the unpinned B-Block lamellae. The unpinned B-block lamellae in a 2x density multiplication are always positioned adjacent to the pinning stripe, meaning that the beads near the substrate may feel the influence of the pinning stripe.[78] This means that while the background region may have a neutral composition, the polymer chains near the substrate for the B-block lamellae feel some level of A-block preference, and then deform

their through-film structure. At the same time, this means that B-block homopolymer will be less likely to interact with the substrate, further changing the through-film structure as it preferentially interacts at the middle and top of the film in the lamellae. For the pinned A-block lamellae, a more neutral background region means that its homopolymer prefers to interact near the bottom of the film even more, partially explaining why the pinned lamellae interfaces have an increase in LER as f_A approaches 0.50 (Figure 4.4).

However what is believed to be the biggest influence to LER and LWR increases with n_{Homo} is the fluctuation of homopolymer concentration through the length of the lamellae. Figure 4.8 shows a colormap depicting homopolymer concentrations through the length of the lamellae where bright yellow is pure A-block homopolymer ($n_{Homo,A} = 1$), dark blue is pure B-block homopolymer ($n_{Homo,B} = 1$) and green are regions where no homopolymer present ($n_{Homo} = 0$) meaning the BCP is present there. The lines drawn over the colormaps represent the BCP interfaces with the red lines representing pinned lamellae interfaces and the black lines representing unpinned lamellae interfaces. Notice from both the top for both $n_{Homo} = 0.10$ (Figure 4.8a & c) and $n_{Homo} = 0.33$ (Figure 4.8b & d) that fluctuations occur in homopolymer through the length of any given lamellae. At low n_{Homo} because the change in pitch caused by the homopolymer's presence is low, these fluctuations do not have as high an effect on the position of lamellae interfaces or their correlation with neighboring interfaces as high n_{Homo} simulations do.

The concentration of homopolymer between any two lamellae should be equal and even throughout the length of the lamellae in order to achieve a low LER. For getting a low LWR, the comparison between homopolymer concentration needs to be done between three lamellae. The lamellae of interest (the one LWR is being calculated on) needs an even distribution of homopolymer throughout its lamellae. In addition its two neighboring lamellae need equal and evenly distributed homopolymer throughout their lamellae in order to achieve a low LWR. These effects, coupled with the segregation of homopolymer to the free surface or the substrate, are believed to be the causes of increased LER and LWR for

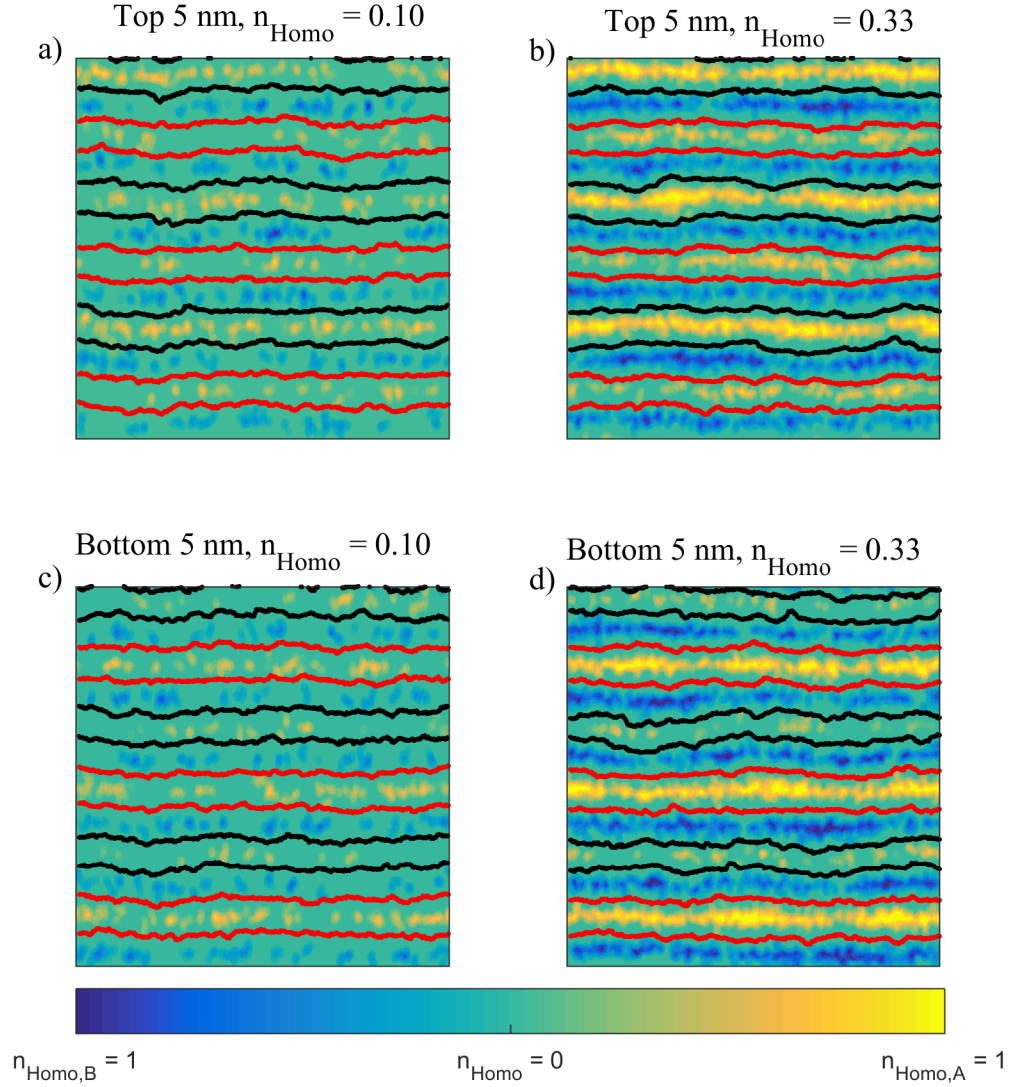


Figure 4.8: Homopolymer concentration ($n_{\text{Homo},i}$) colormaps for the top (a & b) and bottom (c & d) 5 nm of the BCP/homopolymer film simulations for $n_{\text{Homo}} = 0.10$ (a & c) and $n_{\text{Homo}} = 0.33$ (b & d). Lines drawn over the colormaps represent the BCP interfaces, where black lines are interfaces belonging to unpinned lamellae and red lines are interfaces belonging to pinned lamellae.

BCP/homopolymer blends.

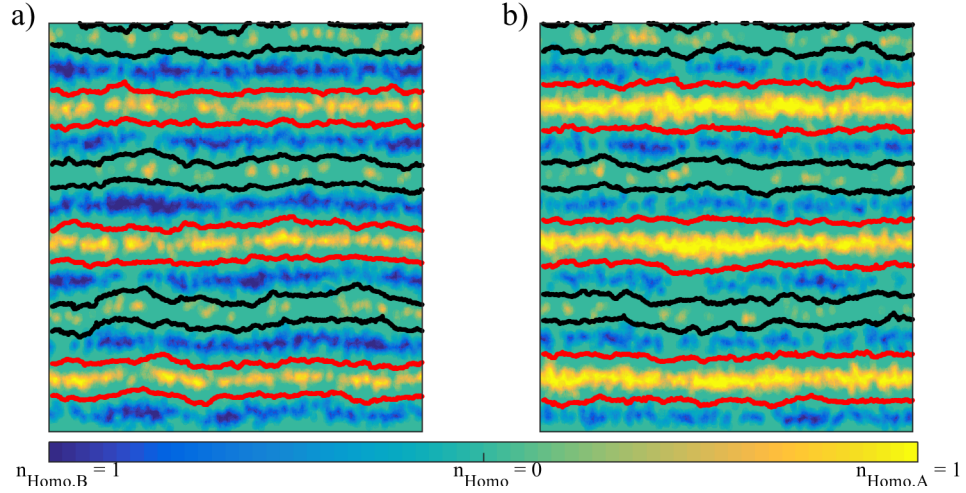


Figure 4.9: Homopolymer concentration ($n_{Homo,i}$) colormaps for the bottom 5 nm of the BCP/homopolymer film simulations for $n_{Homo} = 0.33$ and $f_A = 0.3$ for pinning stripe widths of $0.3 \cdot L_o$ (a) and $0.7 \cdot L_o$. Lines drawn over the colormaps represent the BCP interfaces, where black lines are interfaces belonging to unpinned lamellae and red lines are interfaces belonging to pinned lamellae.

For high n_{Homo} there has been seen a higher impact on LER and LWR with pinning stripe width. Figure 4.9 shows the homopolymer concentration maps for $n_{Homo} = 0.33$ and $f_A = 0.30$ for a pinning stripe width of $0.3 \cdot L_o$ (Figure 4.9a) and $0.7 \cdot L_o$ (Figure 4.9b). As the pinning stripe's width increases from $0.3 \cdot L_o$ to $0.7 \cdot L_o$ the effective underlayer that the B-block (and its homopolymer) feel at the substrate becomes more and more preferential to the A-block. This means that the unpinned B-block lamellae's homopolymer concentration decreases as the pinning stripe width increases. Since the homopolymer concentration in the unpinned B-block lamellae is low, fluctuations along those lamellae do not result in as large of a change in the position of the lamellae's interfaces, lowering LWR and LER. The B-block homopolymer then segregates to the middle and top portions of the film.

The source of these fluctuations in homopolymer concentration are currently unknown but two theories are suggested by the authors. The first is that the fluctuations are caused by the random distribution of homopolymer at its generation. If this is true then it does not

necessarily mean the behavior is unrealistic as the distribution of homopolymer in real in-lab thin film blends may not be completely uniform either. The second theory is that a case of autophobic dewetting is occurring.[111, 112] When a substrate is grafted with polymer chains at a high density and a homopolymer (of the same type) is coated onto this surface, the free-chains may swell the grafted chains and lead to excessive stretching of the grafted chains. This causes an entropic penalty to occur, not to mention the entropic penalty caused by the chains already being partially stretched due to the high grafting density. Combined, this effect can lead to the homopolymer free-chains dewetting on the grafted surface. It's unclear if this effect is occurring in the simulations here, however it is a possibility.

4.3.4 Radius of Gyration

The radius of gyration for homopolymers and individual blocks of the BCP were measured for the simulations. This was done to determine if the conformation of the homopolymer was directed along any particular axis uniformly throughout the thin film. If so, this may affect the degree of stretching in the BCP chain which could have also affected the LER and LWR. The radius of gyration tensor and its eigenvalues were calculated for the chains in the simulation. The eigenvalues for the radius of gyration tensor are a set of three parameters (a , b , and c) that describe the shape of an ellipsoid. If all three values are very close to one another, then the shape is considered spherical while if one value is larger than the other two, then the shape is considered more like a peanut. It should be noted however that the eigenvalues give no information on the direction along which a chain is stretched, here they are ordered least to greatest ($a - c$).

From Table 4.1, it can be seen that both the A/B-homopolymer and the A/B-block of the BCP are stretched primarily along one direction due to their c values being about 5x greater than their b values and more than two orders of magnitude greater than their a values. As stated before, this does not give information on which direction the chain is stretched in. To determine that, the values in the third column of the radius of gyration tensor should be

Table 4.1: Eigenvalues from one simulation with $n_{Homo} = 0.33$, $f_A = 0.3$

-	a	b	c
A-block BCP	0.020	0.073	0.49
A-homopolymer	0.044	0.15	0.84
B-block BCP	0.020	0.074	0.49
B-homopolymer	0.044	0.16	0.83

examined. These values correspond to the vector along which the chain is directed with parameters v_x , v_y , and v_z . The absolute value for each parameter was taken for each chain, and averaged for chains of like-type. An example of a data set is shown in Table 4.2.

Table 4.2: Chain vector parameters from one simulation with $n_{Homo} = 0.33$, $f_A = 0.3$

-	v_x	v_y	v_z
A-block BCP	0.74	0.37	0.36
A-homopolymer	0.46	0.51	0.52
B-block BCP	0.74	0.37	0.37
B-homopolymer	0.46	0.51	0.52

From Table 4.2 it can be seen that both the A and B block of the BCP are directing along the x-axis, the axis perpendicular to the lamellae. This makes sense because that is how they were pre-aligned at the start of the simulation and since there was little morphological change in the system, the chain's primary axis should still be along the x-axis. However the homopolymer chains are not so strongly directed and instead show near-equal values for all three parameters. This means that while the chains are stretched, there are a distribution of directions in which the chains point their primary axis.

4.4 Conclusions

Coarse grained molecular dynamics simulations were used to probe the effects of symmetric homopolymer addition on line edge roughness and line width roughness. While symmetric homopolymer addition can be used to modulate the pitch of BCPs it comes with the adverse effect of deforming the through-film shape of the lamellae. Through-film deformation can affect the transferred pattern into the substrate once one block is removed. Increasing homopolymer concentration (n_{Homo}) was found to increase the LER and LWR for both the top and bottom of the film. This increase in LER and LWR with n_{Homo} is suspected to be due to fluctuations of homopolymer concentration throughout the length of the lamellae. For $n_{Homo} > 0.10$ LER and LWR increases differently in the top and bottom of the film and between pinned and unpinned lamellae, likely due to excessive homopolymer segregation. While LER and LWR for $n_{Homo} < 0.10$ shows little sensitivity to pinning stripe width, at larger n_{Homo} a dependence is seen in unpinned lamellae at either the top or bottom of the film. Variations in the LER and LWR between the surface and substrate can lead to further uncertainty of the integrity of the pattern being transferred.

CHAPTER 5

SYNTHESIS AND SELF ASSEMBLY OF LOW χ BLOCK COPOLYMERS WITH LARGE PERIODICITY

5.1 Introduction

Useful block copolymers (BCPs) are expected to have a high χ to stay above the order-disorder transition but a low enough N to achieve a high density of features. The ITRS has set a defect density target for BCP-DSA of allowing 1 defect every 100 cm². [93] Defects in phase separated BCP materials are detrimental for their lithographic application. A high χ is expected to be troublesome for defect annihilation. As mentioned previously in Chapter 1, dislocation defects annihilate by the terminating blocks of the dislocation penetrating through the opposite block. This increases interactions, causing a penalty, and with a high χ BCP this would be a high penalty. So there's expected to be a growing energetic barrier for defect annihilation for higher and higher χ materials. Conversely, a lower χ material may be useful for decreasing the energetic barrier to defect annihilation. Expanding the library of low χ BCP materials will be helpful in determining the trade off between the energetic barrier and χ .

One application where defect annihilation is crucial is in photonic crystals. Photonic crystals are periodic arrays of features with a separation distance near the scale of the wavelength of visible light and are used to reflect or trap particular wavelengths. [45, 46, 47] Were large defects present in these materials it would adversely affect their performance by loss of light intensity or parts of the light being out of phase with one another. Block copolymers have the advantage of having a natural driving force for defect removal (lowering the number of unlike interactions in the BCP). In this application using large pitch features a high χ would not be as necessary as for lithographic applications. Here a new low

χ material block copolymer, poly(4-tertbutyl styrene) -block- poly(propyl methacrylate)) (PtBS-b-PPMA), was synthesized via anionic polymerization. PtBS-b-PPMA was characterized by gel permeation chromatography (GPC), nuclear magnetic resonance (NMR), and thermogravimetric analysis (TGA). Small angle X-ray scattering (SAXS) was used to characterize the pitch and morphology of PtBS-b-PPMA, as well as determining the χ of the material. A “neutral” underlayer was synthesized using the BCP’s monomers with a benzocyclobutene (BCB) as a crosslinking agent. Thin film BCP samples were prepared, thermally (or solvent) annealed, and then examined via scanning electron microscope (SEM).

5.2 Experimental Methods

Monomers 4-tertbutylstyrene (tBS) and propyl methacrylate (PMA) were purchased from Sigma Aldrich and Alfa Aesar, respectively. All other chemicals were purchased from Sigma Aldrich unless otherwise stated. Unstabilized tetrahydrofuran was purchased from VWR and then run through a solvent purification still prior to use that lowered its water content to 30 - 50 ppm. Monomers were purified as well before use via distillation to remove impurities such as inhibitors and water.

Monomers and diphenylethylene (DPE) were distilled into flame dried flasks. Tert-butylstyrene was added to a flask and underwent three freeze-pump-thaw cycles (FPT) and back-filled with argon before each new cycle. The monomer was then vacuum distilled at 80°C to a flask with vacuum dried dibutyl-magnesium and allowed to stir for 20 minutes. This flask was then distilled yet again into an empty flask and then transferred to a flame dried vial using a cannula for storage and packed under argon. PMA monomer had a slightly different procedure. PMA was added to a flask with calcium hydride, followed by three FPT cycles with a back fill of argon after each cycle, and allowed to stir overnight. The monomer was then vacuum distilled at room temperature into an empty flask. To this flask, the PMA was titrated with trioctylaluminum until the solution turned bright yellow and was allowed to stir for 20 minutes. The monomer was then distilled into a flask, transferred

into a flame dried vial via cannula, and stored under argon. The DPE was placed in a flask and given three FPT cycles with a refill of argon between each cycle. The DPE was then vacuum distilled at 195°C into a flask with vacuum dried n-butyl lithium. Upon stirring in the new flask the DPE turned dark red and was allowed to stir for 20 minutes. After distillation into a new flask, the DPE became colorless, was transferred to a flame dried vial using a cannula, and packed under argon.

A typical polymerization procedure is as follows. Before reactions, a 500 mL reactor was placed in a vacuum oven and exposed to hexamethyldisilazane (HMDS) overnight at 220°C. The reactor was then placed in a glovebox, charged with LiCl, capped by a septum seal, taken out of the glovebox and flame dried three times while having vacuum pulled on the reactor. The reactor was then attached to a purification still and THF dispensed into it followed by one FPT cycle. The reactor was then chilled in an acetone/dry ice bath (-78°C) and allowed to stir. A 1.4M sec-butyl lithium (sec-BuLi) solution in hexanes was then removed via glass syringe from a container stored in a glovebox. A small portion of tBS monomer was added to the now chilled reaction followed by titration using the sec-BuLi. The THF from the distillation still, while exceptionally pure, did still contain some traces of impurities that would react with any added sec-BuLi. The titrated sec-BuLi was added to remove these impurities while the tBS was used as a colored indicator for when the impurities were removed. When the remaining impurities in the THF were cleaned, the reaction remained bright yellow. Sec-BuLi was then added to the stirring solution in the proper quantities as initiator for the polymerization. Tert-butylstyrene monomer was added, turning the solution bright orange, and allowed to stir for 60 minutes. DPE was then added, turning the solution bright red, and allowed to stir for 60 more minutes. The second monomer PMA was added, turning the solution colorless, and allowed to stir for 180 minutes. Finally the reaction was terminated using anhydrous methanol that had been through three FPT cycles. If homopolymer PtBS was found to be present in the BCP a purification procedure was used. Poly(tert-butylstyrene) was found to be soluble in decane

while PPMA was insoluble. When necessary, BCP powders were allowed to stir in decane for two hours and then precipitated out into methanol to the point that PtBS homopolymer in the product was < 15 mol%.

Characterization of the BCP film was done using GPC, NMR, TGA, and bulk SAXS. SAXS was performed on the Xenocs Xeuss 2.0, Line Eraser using a Pilatus 300K SAXS detector. All bulk SAXS samples were annealed at 150°C for 12 hours prior to their measurement. Thin film studies were conducted for the annealing of the BCP films and examined using a Zeiss Ultra60 FE-SEM. Etch contrast between the blocks was done using Lam Research Flex45 Dielectric Etcher (300 mm) and an Ar/O_2 recipe.[113] The Ar/O_2 -based RIE was done in a 300 mm chemically confined capacitively coupled plasma chamber (Lam Research FlexTM Series) under standard low pressure conditions ($< 100\text{mT}$) with 60MHz RF source power supplied from the bottom electrode.

A neutral, random copolymer underlayer was synthesized for PtBS-b-PPMA via radical polymerization using tBS, PMA, and 4-vinylbenzocyclobutene (BCB). BCB was obtained from BOC Science and used as the crosslinking agent. Monomers were run through an alumina column to remove their inhibitors. They were then added to a schlenk flask with AIBN and toluene and stirred at 75°C for 8 hours. The reaction was then cooled to room temperature, quenched via exposure to air, and precipitated in methanol to obtain a white powder. The underlayer was characterized by NMR and its thin films by water contact angle measurements to determine its preference to the homopolymers. Thin films of the underlayer were made by first coating the underlayer on an HMDS primed wafer piece followed by a soft bake at 90°C for two minutes. The film was then baked at 250°C for 15 minutes to crosslink the BCB and then rinsed with PGMEA. Ellipsometry shows typical underlayer thicknesses were 40 - 60 nm. BCPs were coated atop these and then either solvent or thermally annealed.

5.3 Results and Discussion

5.3.1 Synthesis

PtBS-b-PPMA (Figure 5.1) was synthesized via anionic polymerization using the procedure previously mentioned. It was found however that after the polymerization there was a substantial secondary peak in the GPC trace which was determined to most likely be dead PtBS homopolymer. After the polymerization of PtBS, DPE was added and then followed by PMA. During the addition of DPE and/or PMA, it is likely a contaminant entered our system and terminated some of the PtBS homopolymer. The homopolymers and the BCP were mixed with several solvents to find a solvent that was preferential to PtBS but not PPMA. Decane was found to be an appropriate solvent. The BCP was stirred in decane for 3 hours, filtered, and then re-precipitated in chilled methanol. A GPC trace of the BCP showed the removal or lessening in the secondary peak while NMR showed an increase in the peak ratios for PPMA compared to PtBS, supporting the notion that the decane had removed homopolymer PtBS. An example of the final GPC and NMR traces are shown in Figure ??a and ??b, respectively. The range of volume fraction of PPMA calculated from the NMRs was 0.32 - 0.52 for all our samples.

5.3.2 SAXS Characterization

Bulk SAXS was conducted on the BCP samples to determine their morphology, pitch and (for sub-ODT samples) the χ of PtBS-b-PPMA. Bulk samples were prepared by creating a mold for a disk with a 5 mm diameter and a 1 mm thickness. BCP powder was placed in the mold and a few drops of PGMEA was added. The mold was then placed in a vacuum oven at 90°C for 30 minutes. The temperature was then raised to 150°C and it was allowed to anneal for 18 hours. The BCP disk was then peeled off of the aluminum foil and SAXS was performed. Examples of SAXS results are shown in Figure 5.3. The primary peak (q^*) in the SAXS data will have reflections of its position which indicate the morphology of the

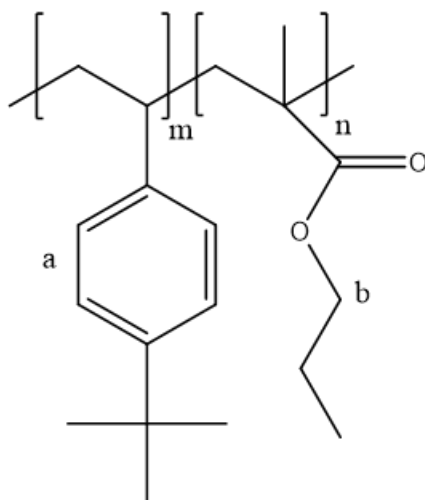


Figure 5.1: Structure of PtBS-b-PPMA. The increase in hydrocarbon groups to each block is expected give a lower χ value for this BCP compared to PS-b-PMMA

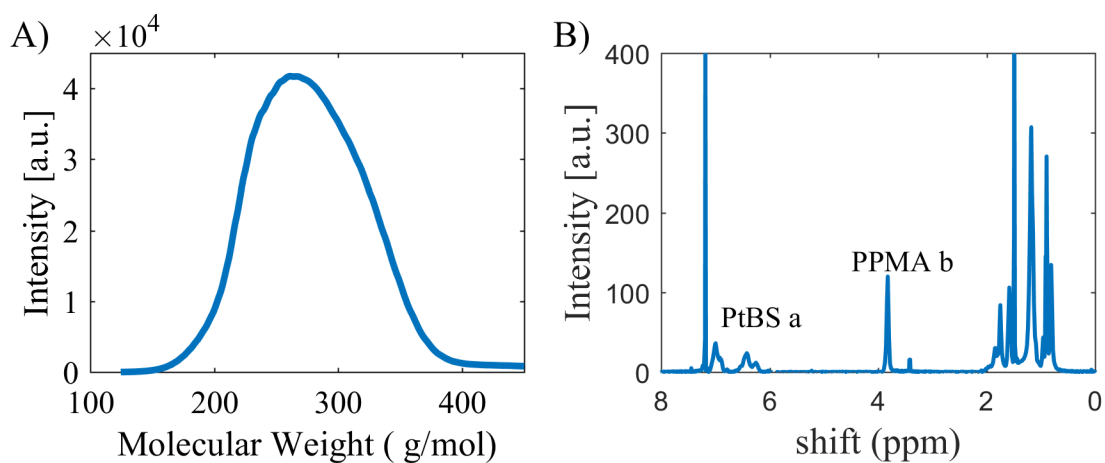


Figure 5.2: A) Example of a GPC trace of PtBS-b-PPMA after washing in decane. B) Example of ^1H -NMR for PtBS-b-PPMA. The aryl protons (4H, a) for PtBS are shown from about 7.25 - 6.0 ppm. The protons on the carbon adjacent to the ester group in PPMA (2H, b) are shown at 3.85 ppm. The ratios of these two can give the mole fraction of each component in the BCP which can then be used to determine the volume fraction.

phase separated features. The position of q^* can be used to determine the pitch of the phase separated features. If the BCP is below the order-disorder transition (ODT), the primary peak may be fit to Leibler theory[26] to determine the χ of the BCP. A BCP may be below its ODT by having a χN that is too small, or by having a thermally sensitive χ and raising the temperature high enough to go below the ODT.

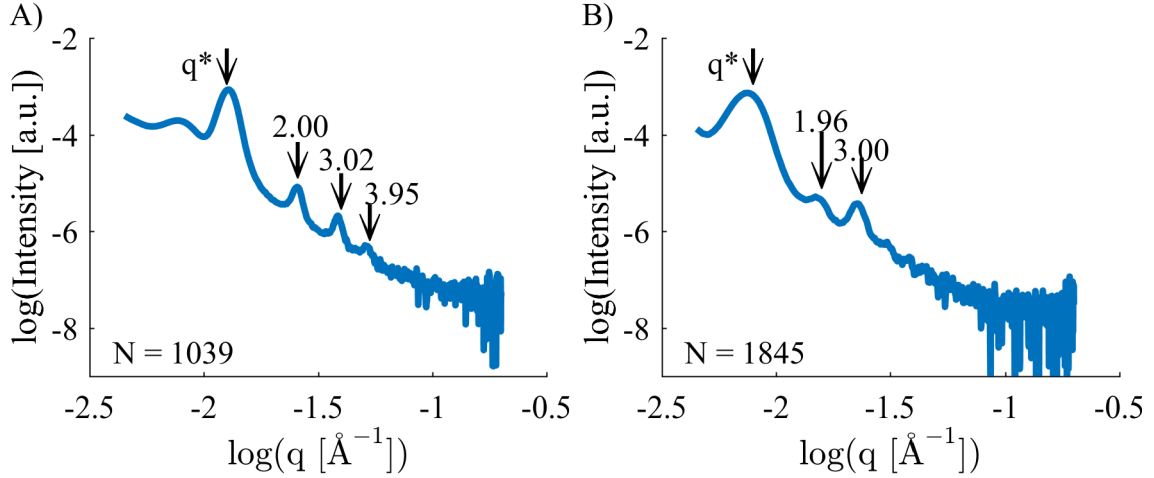


Figure 5.3: Representative SAXS data for PtBS-b-PPMA annealed at 150°C. Both SAXS measurements show a primary peak (q^*) followed by integer reflections of this peak denoting lamellae formation. The pitch of the morphology determined by SAXS for these two BCPs are A) 51 nm and B) 83 nm.

Figure 5.3 shows two examples of our SAXS measurements, both of which have near-integer reflections of their primary peak. This means that the BCPs phase separated into a lamellae morphology. The position of q^* can be used to determine L_o ($= 2\pi/q^*$), which was found to be 51 nm and 83 nm, respectively, for Figure 5.3a and 5.3b. From this data, we could estimate what the χ of PtBS-b-PPMA is by comparing its L_o to the L_o for PS-b-PMMA. Equation 5.1 shows the relation for L_o for a given BCP, where a_{BCP} is the statistical segment length for the BCP

$$L_o \approx a_{BCP} \cdot \chi^{\frac{1}{6}} \cdot N^{\alpha} \quad (5.1)$$

The exponent α is dependent on whether the BCP is in the strong segregation regime ($\chi N > 40$, $\alpha = 2/3$) the intermediate segregation regime ($40 > \chi N > 10$, $\alpha = 0.8 - 1.0$), or the weak segregation regime (near the ODT, $\alpha = 1/2$). A recent paper showed the self-assembly of PS-b-PMMA for $N = 1040$ having a $L_o = 48$ nm.[114] This is similar to PtBS-b-PPMA having an $N = 1039$ and a $L_o = 51$ nm. However the statistical segment lengths are quite different from these two BCPs. The average a_{BCP} value for PS-b-PMMA is 0.68 nm[115], while the average a_{BCP} for PtBS-b-PPMA is 0.77 nm.[62, 116] This increase in statistical segment length for PtBS-b-PPMA compared to PS-b-PMMA suggests that for the same L_o , using Equation 5.1, PtBS-b-PPMA should have a lower χ than PS-b-PMMA. Using Equation 5.1, the χ of PtBS-b-PPMA is estimated as 0.027.

Four low N samples of PtBS-b-PPMA were made in order to determine the χ using SAXS and Leibler theory. Figure 5.4 shows the SAXS data of the four samples as well as their N and the full width half max (FWHM) of their primary peaks. Immediately the lack of reflections of q^* suggests we are near or below the ODT with each sample. Previous research has shown that by measuring the FWHM of q^* as a function of temperature, the ODT can be found by a sudden increase in the FWHM, a broadening of the primary peak.[62, 61] As the temperature changes, χ decreases, lowering χN until the ODT is passed. Determining χ can be done by varying N . From Figure 5.4 it can be seen that $N = 158$ has such a broad primary peak that the peak itself is barely identifiable. This means that $N = 158$ is definitely below the ODT but perhaps too far below it to get χ . Looking at $N = 367$, 458, and 569 we can see a clear change in the shape of q^* and in the FWHM of their peaks. Between $N = 367$ and 458, the FWHM decreases by almost a factor of 3x, while between $N = 458$ and 569 the FWHM stays relatively constant. This suggests that the ODT at 150°C for the BCP is somewhere between $N = 367$ and 458. Figure 5.5 illustrates this more clearly with the FWHM of each sample's primary peak. At high molecular weights the FWHM stays relatively constant at a lower value. This is in contrast to the significantly higher FWHM at $N = 367$ when the BCP disorders.

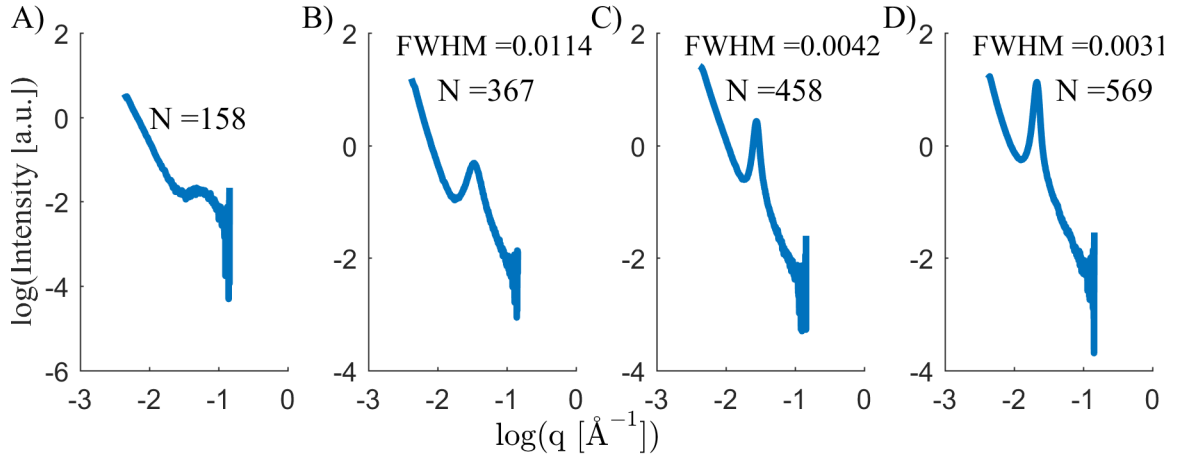


Figure 5.4: SAXS data of samples annealed at 150°C approaching the ODT, denoted by a sudden drop in the full width half max (FWHM) of the primary peak.

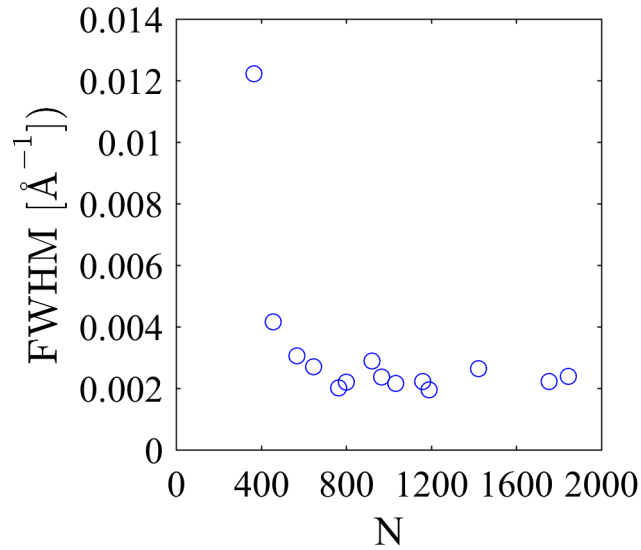


Figure 5.5: Plot of the full width half max of the primary peak (q^*) for each sample. The sudden broadening of the primary peak when lowering to $N = 367$ denotes the ODT has been crossed.

Figure 5.6 shows the Leibler theory fit for $N = 367$ annealed at 150°C . To see the equations used for the fit, the reader is directed to Appendix A. The model was fit to the data using χ , a_{BCP} , and K (a proportionality constant) as adjustable parameters. From this, χ and a_{BCP} were fit to be 0.027 and 0.75 nm, respectively. Despite the statistical segment length being a little off from the average of the literature values[62, 116] it still agrees with literature quite well for the expected lengths for the homopolymers. The fitted χ also shows good agreement with our previous estimation using Equation 5.1 for PtBS-b-PPMA and PS-b-PMMA. It is worth mentioning, that a sample of PS-b-PMMA just below its expected ODT was also run through this SAXS tool and fit using Leibler Theory. In this fit, we found that for PS-b-PMMA (10,100-b-9,700) χ and a_{BCP} were fit as 0.048 (annealed at 150°C) and 0.72 nm, respectively. This χ value for PS-b-PMMA is in close agreement with the literature (0.038 at 150°C).[61]

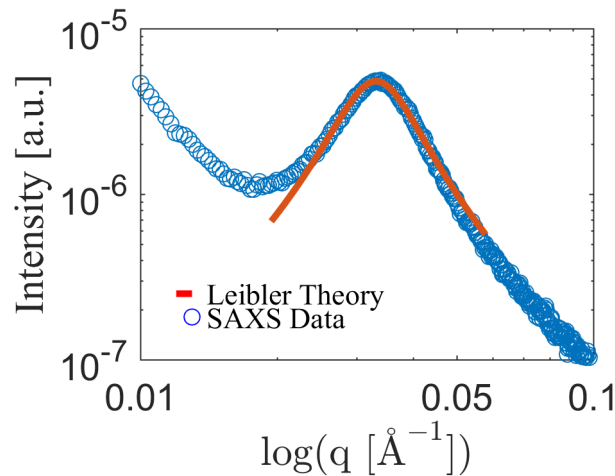


Figure 5.6: SAXS data of $N = 367$ PtBS-b-PPMA annealed at 150°C fit by Leibler theory (line). Here χ and a_{BCP} were adjustable parameters found to be 0.027 and 0.75 nm, respectively

All remaining samples showed SAXS reflections that were integer whole numbers of q^* , meaning they all formed a lamellae morphology. Figure 5.7 shows our samples with $N > 367$ superimposed over the mean field phase diagram. With the exception of two points, the mean field phase diagram would suggest that all of our samples would form the lamellae

morphology. Surprisingly, the point at $f_{PPMA} = 0.32$ formed lamellae as well, where the mean field phase diagram would have predicted a cylindrical phase. This highlights the fact that the mean field phase diagram is for an ideal polymer (symmetric in volume fraction, density, statistical segment length, etc) while real polymers are typically asymmetric in one or more of these aspects. These asymmetries can cause changes to the shape of the phase diagram for a BCP, sometimes even excluding phases that would be possible in symmetric cases.[27, 117, 81] PtBS-b-PPMA is very symmetric in statistical segment length but is asymmetric in its density ($\rho_{PPMA} / \rho_{PtBS} = 1.12$).

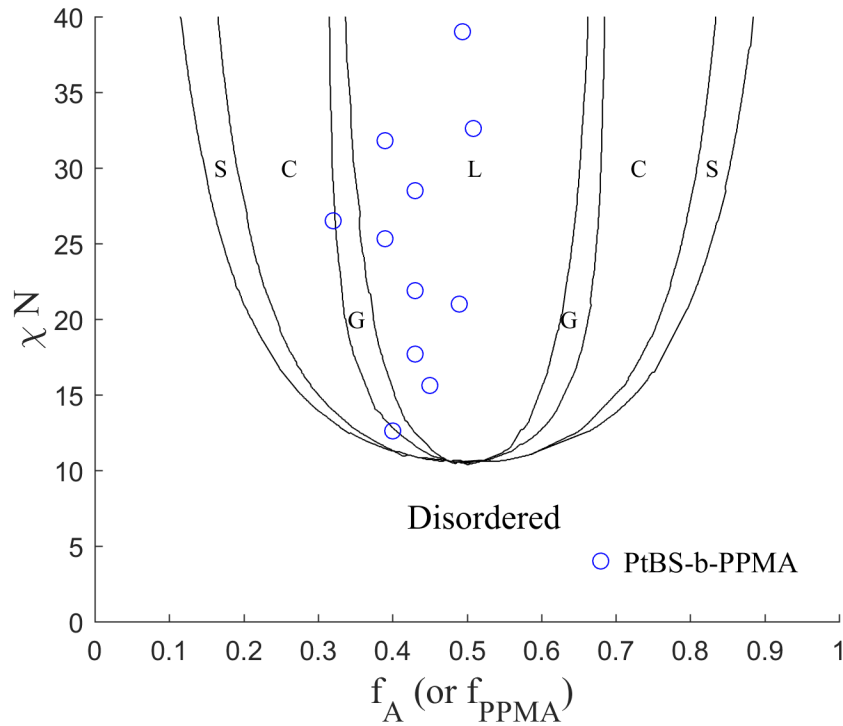


Figure 5.7: Plot of experimentally determined lamellae forming PtBS-b-PPMA (o) superimposed over the mean field phase diagram for an ideal BCP.

For BCPs, L_o scales with N^α , where α varies depending on the degree of segregation. At roughly $\chi N < 12$ the BCP is considered to be in the weak segregation regime with $\alpha = 0.5$. [26, 33] At the range $12 < \chi N < 40$ the BCP is in the intermediate segregation regime. In this regime the value of α can vary between 0.8 - 1.0. [118, 119] At $\chi N > 40$ the BCP

is in the strong segregation regime with $\alpha = \frac{2}{3}$. [34] Figure 5.8 shows the SAXS determined pitch for PtBS-b-PPMA samples with a $\chi N > 20$ and a best fit trend line (dashed line). The samples show an α scaling of 0.948. The largest N sample had a $\chi N = 50$, yet there does not appear to be a deviation from the near 1.0 α scaling towards the expected $\alpha = \frac{2}{3}$.

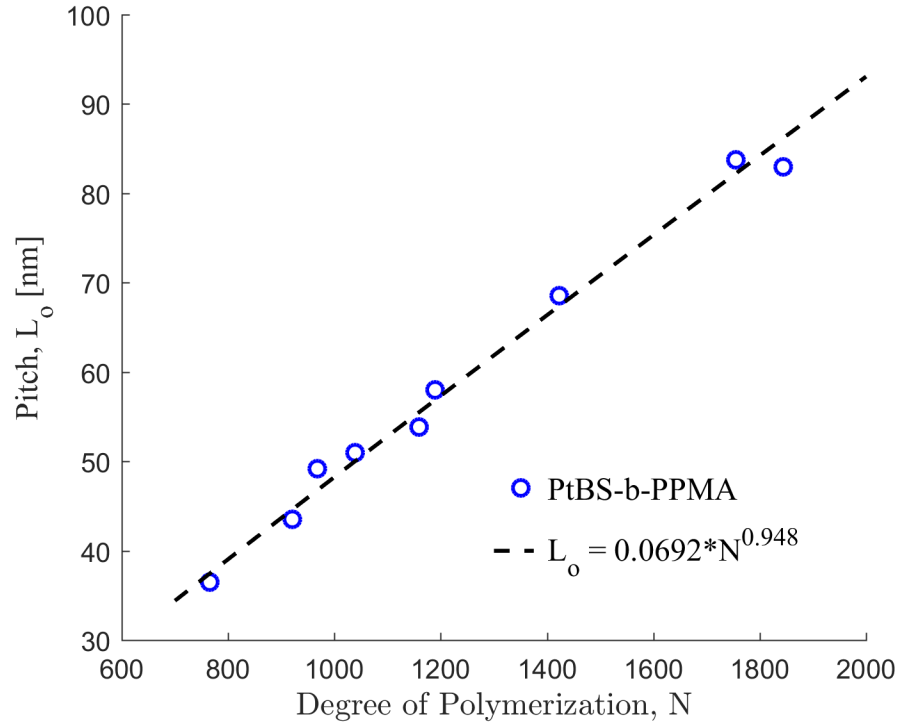


Figure 5.8: Plot of L_o versus the degree of polymerization for PtBS-b-PPMA samples (o) with a $\chi N > 20$ and a corresponding trend line (dashed line) to the data.

5.3.3 Thin Film Studies

Before the annealing conditions of the BCP were investigated, an etch study was done on the homopolymers to determine the etch contrast between PtBS and PPMA. Silicon wafers were primed with HMDS and then homopolymers of PtBS and PPMA were coated on each wafer. Figure 5.9 shows the results of the etch study conducted by Lam Research Corporation using an Ar/O₂ recipe. [113] PtBS and PPMA have an etch contrast of about 2.3, meaning that PPMA etches 2.3x faster under these conditions than PtBS. This makes

potential lithographic applications of the BCP possible since these applications require one block to be etched away and the other block to remain as an etch mask for the substrate.

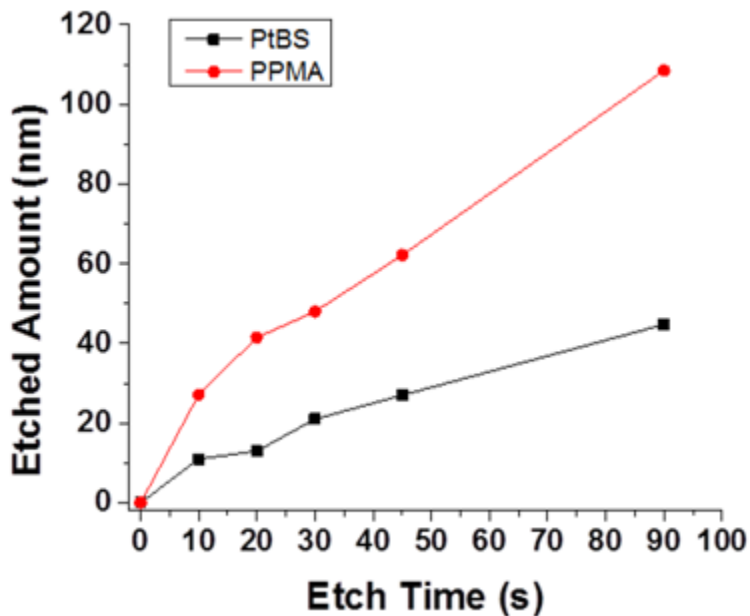


Figure 5.9: Ar/O₂ etch study provided by Lam Research Corporation for PtBS (black) and PPMA (red) showing an etch contrast of 2.3 between the blocks.

Annealing of a block copolymer film has commonly been done by thermal and/or solvent annealing. In thermal annealing the BCP's temperature is raised above the glass transition temperature (T_g) of the two homopolymer blocks to increase chain mobility and phase separation kinetics. In solvent annealing, the BCP film is swollen with a neutral solvent in order to lower the effective T_g of the two homopolymer blocks to increase chain mobility. Both annealing conditions were explored for PtBS-b-PPMA. First though, a thin film of the BCP had to be coated atop a neutral underlayer to ensure one block does not preferentially wet the substrate it was coated on. To do this, a crosslinkable, random copolymer of PtBS, PPMA and BCB was created. The water contact angle of PtBS and PPMA was found to be 86° and 104° while the random copolymer underlayer had a water contact angle of 95°. At this contact angle the underlayer can be considered “neutral”, since this value is the median of the two homopolymer water contact angles. After crosslinking and rinsing the

underlayer, 40 - 50 nm of underlayer remained and the BCP was coated atop this. Atomic force microscopy probing of the final underlayer showed the thickness oscillated by about 1 nm over the surface. It is worth noting that the underlayer here is thicker than would be used for pattern transfer of the self-assembled pattern. This was done for experimental convenience in obtaining fingerprint patterns.

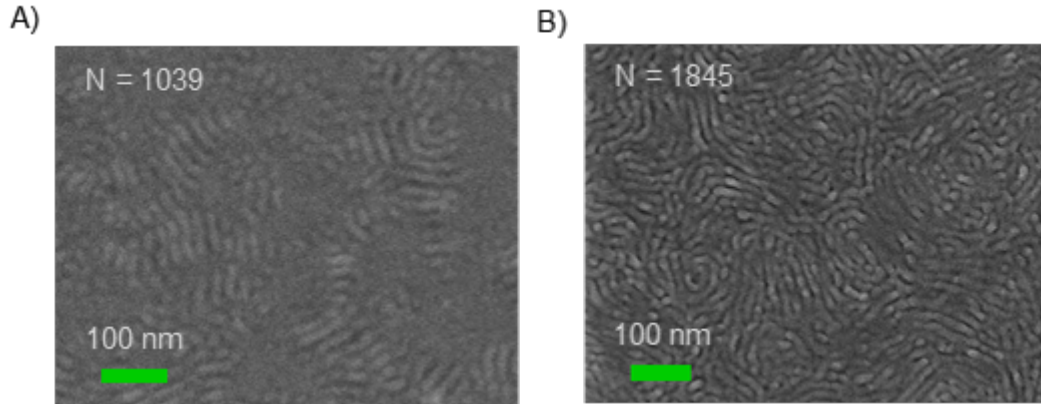


Figure 5.10: SEMs of as-cast PtBS-b-PPMA film surfaces spin coated from PGMEA solutions for A) $N = 1039$ with an $L_o = 55$ nm and B) $N = 1845$ with an $L_o = 83$ nm.

Thermal annealing was attempted first. Block copolymer thin films were coated from PGMEA onto the underlayer with thicknesses ranging from $1L_o$ - $1.75L_o$, followed by a two minute post-apply bake at 90°C . Films were then placed inside of a vacuum oven and thermally annealed while under vacuum. Figure 5.10 shows the state of the surface of the PtBS-b-PPMA film after spin coating from PGMEA for two samples. The as-cast state for $N = 1845$ (Figure 5.10B) shows swollen long lamellae while for $N = 1039$ (figure 5.10A) only small patches of growing perpendicular lamellae grains can be seen. Initially, due to the high N , it was expected high temperatures and long times would be needed to achieve long range order. Films were placed in the vacuum oven at 200°C for 60+ hours, only to find what appeared to be de-wetted droplets of BCP on the surface of the film. Thermogravimetric analysis (TGA) was then performed on the BCP to find that the PPMA block was significantly thermally sensitive at temperatures above 180°C . Bulk PPMA was placed

under several different temperatures for long periods of time and the rate of mass loss measured. The decomposition of PPMA is suspected to be due to a combination of main chain scission and ester decomposition.[120, 121] Assuming a zeroth order decomposition of PPMA, the activation energy can be determined using the Arrhenius Equation. The activation energy for PPMA decomposition was found to be 73.4 kJ/mol and the pre-exponential was found to be 6.99 kg/min. Figure 5.11 shows the expected loss of PPMA volume from the BCP over time from a BCP with an initial PPMA content of 48 vol%. It was found that 150°C is a safe temperature to anneal at.

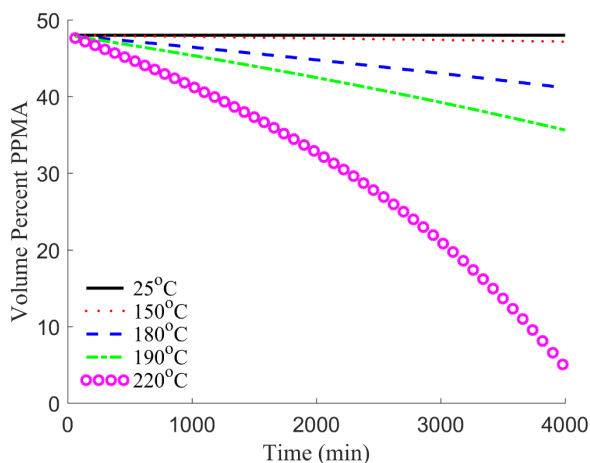


Figure 5.11: From thermogravimetric analysis the degradation of PPMA was found over time and estimated for BCPs at various temperatures.

Figure 5.12 shows the progression of the $N = 1039$ (BCP film thickness = $1.3L_o$) sample as it was annealed at 150°C. After 18 hours, large areas of fingerprint lamellae could be seen over the film. Strangely, however, at the intermediate time of 6 hours the surface morphology looks quite different compared to 18 hours. At 6 hours, the surface of the film is covered by what appears to be cylindrical morphology. Due to the lack of through-film knowledge however we will refer to these as “dots” instead. Previous studies of PS-*b*-PMMA have shown that this structure can be an intermediate for lamellae forming BCPs on the way to lamellae.[122, 123] The SAXS data from Fig. 3 bolsters this idea since it

shows this material as forming lamellae with multiple reflections of q^* . It is suggested that due to the high N the BCP has low mobility, and orders itself with its nearest neighbors first and then proceeds to form the correct morphology. Despite producing fingerprint lamellae, 18+ hour annealing times are not very attractive for the BCP's ultimate application.

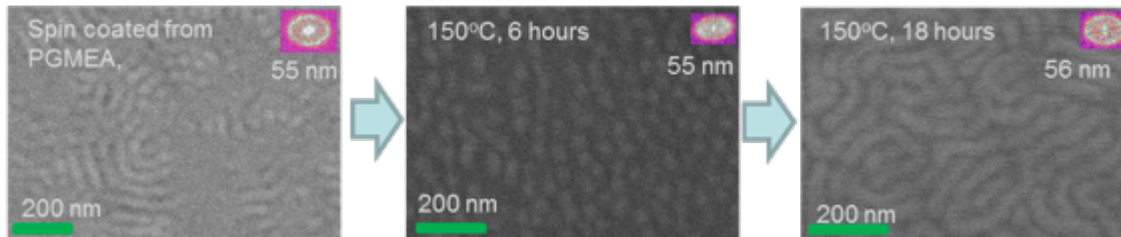


Figure 5.12: Thermal annealing of PtBS-b-PPMA ($N = 1039$, Film Thickness = $1.3L_o$). Lamellae go through a cylinder/dot formation before forming fingerprint lamellae.

Solvent annealing has shown to be an effective way to increase the mobility of polymer chains by depressing their T_g . The difficulty however is in finding a solvent that is “neutral” to both blocks and will not preferentially swell one of them. If a solvent is too preferential, it could significantly change the effective volume fraction of the block it swells and, in the extreme, change the morphology the BCP separates into. As a rough estimate, solubility parameters of the BCP and its potential neutral solvents are used to determine if a solvent is a neutral solvent. Table 1 shows the literature values for the solubility parameters of PtBS, PPMA and the solvents used for annealing here.[124, 125, 126, 127] Ideally, Hansen solubility parameters should be used over Hildebrand. Hansen’s parameters try to account for the dispersion (δ_D), polar (δ_P), and hydrogen bonding (δ_H) interactions that play a role in solubility while Hildebrands parameter groups all of these factors together.

Unfortunately, PtBS and PPMA only have Hildebrand parameters in the literature. Compared to PS and PMMA and accounting for their increased content of hydrocarbons, it is assumed the majority of PtBS-b-PPMA interactions are due to dispersion forces and not polar or hydrogen bonding, suggesting that using Hildebrand parameters will be useful here. The solvents chosen as candidates for neutral solvents for this initial study into

Table 5.1: Solvent/Polymer Solubility Parameters from Literature

Polymer/Solvent	Hildebrand δ_i [MPa ^{0.5}]	Hansen δ_D [MPa ^{0.5}]	Hansen δ_P [MPa ^{0.5}]	Hansen δ_H [MPa ^{0.5}]
PtBS	16.6	-	-	-
PPMA	17.9-20.0	-	-	-
THF	19.4	16.8	5.7	8.0
PGMEA	19.3	15.6	5.6	9.8
Toluene	18.3	18.0	1.4	2.0
Cyclohexane	16.8	16.8	0.0	0.2
Hexane	14.9	14.9	0.0	0.0

solvent annealing of PtBS-b-PPMA were propylene glycol methyl ether acetate (PGMEA), tetrahydrofuran (THF), toluene, cyclohexane and hexane. PGMEA was chosen due to the presence of lamellae in the as cast state as shown in Figure 5.10. The presence of these lamellae imply that PGMEA may be close to a neutral solvent. Due to the range of solubility parameters for PPMA in the literature, toluene was chosen as a potential “neutral” solvent if δ_{PPMA} had a value of 20.0 (the largest value found in literature for PPMA). Here, “neutral” is described as being close to the average of the solubility parameters of PtBS and PPMA. Hexane and cyclohexane were chosen due to their primary interactions being from dispersion forces. While hexanes solubility parameter (14.9) is outside the range of δ_{PtBS} and δ_{PPMA} (16.6 - 20.0) cyclohexanes is just inside that range (16.8). Tetrahydrofuran was chosen due to its ability to dissolve both homopolymers as their reaction solvent during synthesis.

Solvent annealing was done by placing thin films into a metal O-ring sealed chamber and pulling a vacuum down to 0.1 torr. Solvent vapor was evaporated at room temperature into a vessel 23x larger in volume than the one holding the thin film sample. Annealing was performed under a solvent activity of 0.7, 0.8, and 1.0. The activity of a pure solvent vapor is defined as the pressure of the solvent vapor divided by the saturation pressure of

the solvent vapor at the temperature of the vessel holding the vapor. All three conditions produced the same morphology for these solvents. At a given time, a pneumatic valve would open between the chambers, pushing the solvent vapor into the sample chamber and in contact with the sample. After a set amount of time, the valve would close, the sample chamber would be pumped down, and the sample removed and baked at 90°C for 5 minutes.

Figure 5.13 shows the results after long annealing times while in contact with each solvent. After 60 minutes of annealing under THF (Figure 5.13E), the BCP seems to have formed a cylindrical morphology, suggesting this was not a neutral solvent. Toluene (Figure 5.13B), hexane (Figure 5.13C) and cyclohexane (Figure 5.13D) formed mixtures of lamellae and cylinders, suggesting these solvents are still preferential to one of the blocks. PGMEA (Figure 5.13A) however showed a significantly different result compared to the other solvents. After 360 minutes, PGMEA annealed films showed mostly fingerprint lamellae at their surface with very few cylinders present. This is odd because based on Table 1, the solubility parameters would not suggest that PGMEA is a good solvent for this BCP given its relatively high δ_P and δ_H . This highlights the fact that solubility parameters are an estimation, and a more accurate model is still needed for solvent preference to polymers. Future studies will look at the effect of directly swelling homopolymer thin films of the blocks in the BCP with each solvent and measuring their uptake via ellipsometry for a more direct comparison of solvent preference.

5.4 Summary & Conclusions

A new block copolymer, PtBS-*b*-PPMA, was synthesized and found to have a $\chi = 0.027$, two-thirds the value of PS-*b*-PMMA. The BCP was observed to phase separate into lamellae fingerprint patterns with L_o ranging from 20 - 83 nm. Thermal annealing shows that at 150°C after 18 hours lamellae fingerprint patterns can form in the thin film. PPMA however was found to be a thermally sensitive block above 180°C, a drawback for thermal annealing

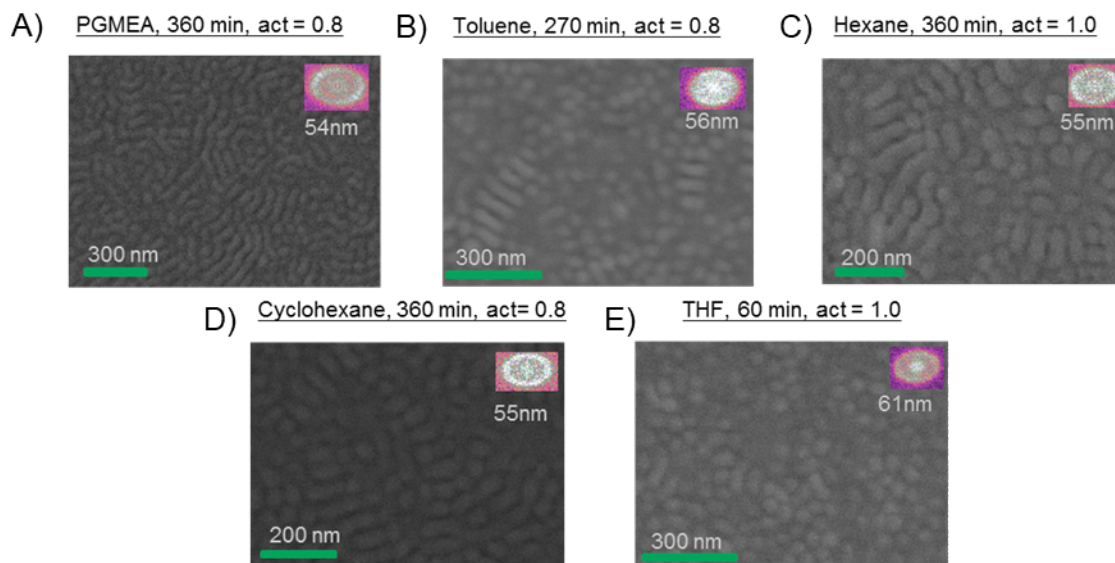


Figure 5.13: Solvent vapor annealing of PtBS-b-PPMA, $N = 1039$. The solvent choice, exposure time, and activity of the solvent (act) are listed above each image. From these solvents, PGMEA appears to be the most 'neutral' by virtue of having the least density of cylinders at the surface.

of the BCP since PtBS has a T_g of 135°C . Initial solvent annealing experiments point to PGMEA as being a possible neutral solvent for the BCP. Further experimentation into the annealing conditions of this BCP will be needed. Whether a single solvent or a mixture of solvents is needed, the BCP will probably need to undergo solvo-thermal annealing to anneal out defects due to the large N needed for phase separation to occur.

CHAPTER 6

SYNTHESIS AND SELF-ASSEMBLY OF HIGH- χ PTBS-B-PHEMA

6.1 Introduction

DSA of BCPs ideally is meant as an extension of optical lithography. Optical lithography would be used to pattern the underlayers necessary to direct the self-assembly of the BCP, and the BCP's natural pitch would be small enough to make the patterns that conventional optical lithography could not. As of 2017, 10 nm node chips have become commercially available, meaning that the smallest printed feature on the chip is 10 nm in width. Future expected node sizes are 7 nm and 5 nm, meaning the feature to feature distance will need to be 14 nm and 10 nm, respectively. To reduced the pitch between their features, BCPs require a low N yet a high enough χ such that the product χN remains above the ODT.

In order to achieve this considerable amounts of research have been conducted to increase the library of high- χ BCPs.[88, 22, 63] Ideally, a "high- χ " BCP will have a χ larger than that of PS-b-PMMA ($\chi = 0.03 - 0.04$) [61], both blocks will have relatively low T_g s, and the BCP will not require a topcoat to form perpendicular features at its surface. A topcoat is a neutral layer at the free surface that is needed when one block has such a strong interaction with the free surface, that it forms a thin layer near the free surface covering possibly perpendicular features beneath it.[50] A topcoat is unattractive due to the increased number of processing steps to coat and remove the layer at the surface. Previously it was observed that polystyrene-block-poly(hydroxyethyl methacrylate) (PS-b-PHEMA) was able to form 15nm pitch lamellae, visible via SEM without the need of a topcoat. [24]. This chapter will describe the synthesis, characterization, and self-assembly of poly(tertbutyl styrene)-block-poly(hydroxyethyl methacrylate) (PtBS-b-PHEMA) as a new high- χ BCP. SAXS will be used to discern the pitch of various molecular weight sam-

ples and AFM and SEM will be used to image thin film samples.

6.2 Experimental Materials and Procedures

Monomers tertbutyl-styrene (tBS) and hydroxyethyl methacrylate (HEMA) were purchased from Sigma Aldrich. Tertbutyl-dimethylsilyl chloride (TBDMS-Cl), 4,4'-dinonyl-2,2'-dipyridyl(dNbpy), ethyl bromoisobutyrate (EBiB), copper chloride, toluene, sec-butyl lithium (sec-BuLi), lithium chloride, tertbutyl ammonium fluoride, diphenylethylene (DPE), n-butyl lithium, dibutyl magnesium, hexamethyldisilazane, acetoxystyrene, propylene glycol monomethyl ether acetate (PGMEA), dimethyl formamide (DMF), and calcium hydride were also purchased from Sigma Aldrich. Unstabilized tetrahydrofuran (THF), dichloromethane, methanol, acetone, ethyl acetate, and magnesium sulfate was purchased from VWR and then run through a solvent purification still prior to use that lowered its water content to 30 - 50 ppm. Alumina (80-200 mesh) and triethylamine were purchased from Fischer Chemical. Hydrochloric acid (12 M) was purchased from Acros Organics. The crosslinking agent for underlayers, 4-vinylbenzocyclobutene (BCB), was purchased from BOC Science. Deuterated chloroform and deuterated dimethyl sulfoxide (DMSO) for NMR solvents were purchased from Cambridge Isotope Laboratories. Triphenyl sulfonium hexafluoroantimonate (TPS-SbF₆) was purchased from Midori Kagaku, Ltd. All materials are used as received unless otherwise stated.

6.2.1 Protection of HEMA Monomer

Prior to synthesis a TBDMS protecting group was placed on the HEMA monomer to shield the propagating chain from the hydroxyl of HEMA (Figure 6.1). HEMA monomer was first run through an alumina column to remove its inhibitor and then dried. A typical protection reaction used 13.5 mL of HEMA, 20 mL TEA (1.29 molar equivalent to HEMA), 20.2 grams of TBDMS-Cl (1.21 molar equivalent to HEMA), and 100 mL of dichloromethane. The reaction was allowed to stir for 24 hours at room temperature. Afterwards the product

was washed with a 3% solution of hydrochloric acid three times, followed by three washings with deionized water. The solution was then poured over magnesium sulfate and dried on the rotavap leaving behind a dark yellow product. Figure 6.2) shows the NMR of the final product in deuterated chloroform with peaks at 0.15 (h₁, 6 protons) and 0.98 ppm (h₂, 9 protons) denoting the dimethyl and tertbutyl protons of TBDMS, respectively, while the alkyl peaks of HEMA are seen at 4.2 ppm (h₃, 2 protons) and 4.0 ppm (h₄, 2 protons).

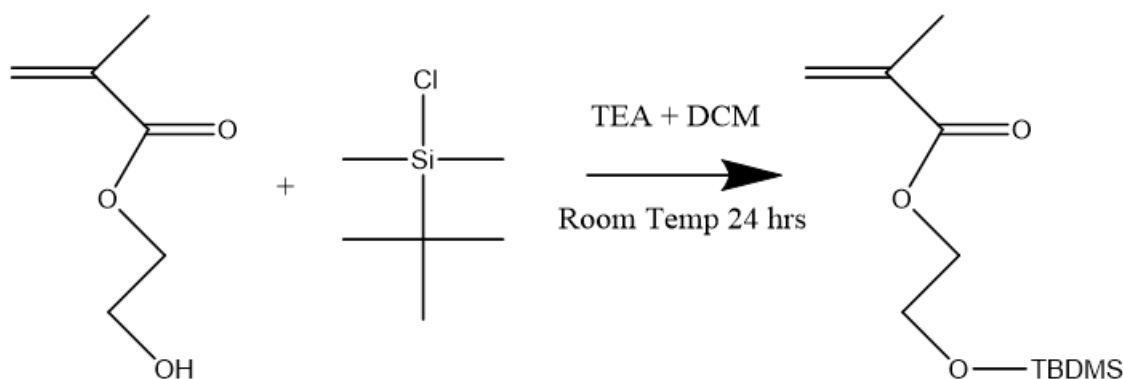


Figure 6.1: Scheme for the protection reaction of HEMA monomer by TBDMS.

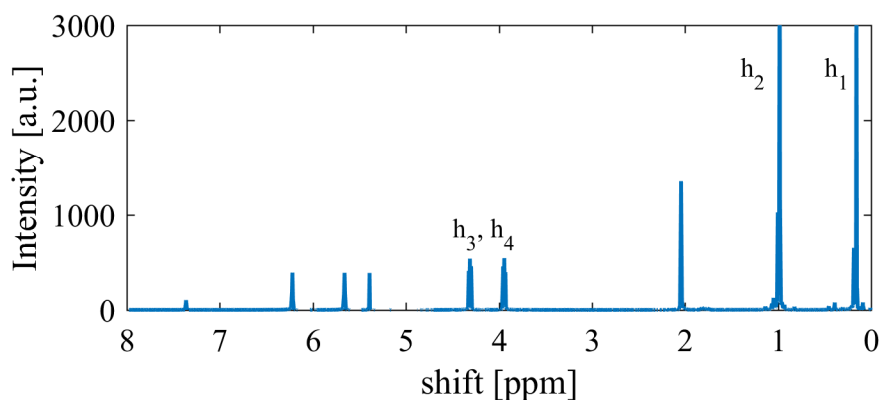


Figure 6.2: NMR of protected HEMATBDMS.

6.2.2 Atom-Transfer Radical-Polymerization of PtBS-b-PHEMATBDMS

For atom-transfer radical-polymerization (ATRP) of PtBS-b-PHEMA, tBS was run through an alumina column prior to use to remove inhibitor. First, the HEMATBDMS monomer

was polymerized using the following procedure. HEMATBDMS (4.74 g), dNbpy (390 mg), toluene (2 mL) and EBiB (67.0 μ L) were placed inside of a 50 mL schlenk flask with a small stir bar. Next copper chloride (45 mg) was added and the flask was sealed with a septum seal and quickly placed in liquid nitrogen. Three freeze-pump-thaw cycles were given to the flask, backfilling with after each thaw. The flask was then placed in an oil bath at 75°C stirring for three hours. After, the product was quenched in an ice bath and opened to the air. The product was then dissolved with a small amount of dichloromethane, run through an alumina column to remove the copper chloride, dried, and then precipitated in cold methanol. The product was then allowed to dry in a vacuum oven overnight at 50°C.

A typical procedure for adding the second block, tBS, is as follows. PHEMATBDMS (3.10 g), tBS (2.00 g), dNbpy (390 mg) and toluene (9 mL) were added to a 50 mL schlenk flask with a stir bar. Next copper chloride (45 mg) were added to the flask and it was immediately septum sealed and placed in a liquid nitrogen bath for three freeze-pump-thaw cycles. The flask was then placed in an oil bath at 125°C for 24 hours. Afterwards the reaction was quenched in an ice bath and opened to the air. The product was run through an alumina column to remove the copper chloride, dried, and then precipitated in cold methanol. Finally the product was dried in a vacuum oven overnight at 50°C. Typical final polydispersities (PDI) of BCPs via this method were around 1.3. Due to this, only a few samples were made via ATRP, with the majority being synthesized via anionic polymerization.

6.2.3 Anionic Polymerization of PtBS-b-PHEMATBDMS

Anionic polymerization requires stricter control over the level of impurities in its reactants. A typical procedure for monomer purification is as follows. All glassware mentioned here are 100 mL round bottom flasks and, prior to being filled, have been flame dried and back-filled with argon three times. Into a distillation flask with a small stir bar, 4 mL of 1.0M dibutyl magnesium in heptane was injected. A vacuum was then pulled on the flask to

remove the heptane. The tBS monomer (10 mL) was taken from its container, as is, and placed into a dry, clean distillation flask. The flask was given three freeze-pump-thaw cycles, backfilling with argon after each thaw. The tBS monomer was then vacuum distilled at 80°C and 0.1 torr pressure into the flask with the dry dibutyl magnesium and allowed to stir for thirty minutes. Afterwards the tBS monomer was vacuum distilled from the dibutyl magnesium flask and into a clean, dry flask. The purified monomer was then transferred to a flame dried, septum sealed vial for storage using a cannula and packed under argon. Purified tBS monomer was then stored in the fridge.

HEMATBDMS was purified using a slightly different procedure. HEMATBDMS (12 mL) was added to a flask with calcium hydride (about 2 g) with a small stir bar and allowed to stir overnight. The monomer was then vacuum distilled at 90°C and 0.1 torr into a clean, dry flask. The monomer was then transferred to a flame dried, septum sealed vial for storage using a cannula and packed under argon. Purified HEMATBDMS monomer was then stored in the fridge.

Prior to purification of diphenylethylene, 1.0M n-butyl lithium in heptanes (about 3 mL) was injected into a dry, clean flask with a stir bar and vacuum dried of its heptane. Diphenylethylene (15 mL) was then injected into a dry, clean flask and given three freeze-pump-thaw cycles, backfilling with argon after each thaw. The diphenylethylene was then vacuum distilled at 95°C and 0.1 torr into the flask with n-butyl lithium. After stirring for thirty minutes a dark red solution was present, signifying low levels of impurities. The diphenylethylene was then vacuum distilled into a clean, dry flask, transferred into a flame dried septum sealed vial, and packed with argon. Purified diphenylethylene was stored in the freezer.

A typical anionic polymerization procedure for PtBS-b-PHEMATBDMS is as follows. A 500 mL flask with a stir bar is placed in a vacuum oven and exposed to hexamethyldisilazane overnight at 220°C. The reactor is then placed into a glovebox, charged with a small amount of lithium chloride, capped with a septum seal and removed from the glovebox.

The flask is then flame dried three times under vacuum, backfilling with argon between each cycle. Next, the reactor is attached to a purification still and THF is dispensed into it followed by one freeze-pump-thaw cycle. The reactor is then chilled in an acetone/dry ice bath (-78°C), allowed to stir, and backfilled with argon. A 1.4M sec-Buli solution in hexanes was then removed via glass syringe from a container stored in the glovebox. The sec-BuLi was then added to the chilled THF, turning the solution bright yellow. The color mentioned here is used to denote the presence of specific anions and thus is an indication of proper purity levels. The purified tBS monomer was then added via syringe, turning the solution bright orange, and allowed to stir for 60 minutes. Next the diphenylethylene was added via syringe, turning the solution bright red, and allowed to stir for 60 minutes. Finally, purified HEMATBDMS was added via syringe, the solution turned colorless, and was allowed to stir for 180 minutes. During this time methanol was placed into a small vial and given three freeze-pump-thaw cycles. The methanol was then added to the solution at the end of the reaction to terminate the propagating chain. Typical PDIs of BCPs using this method were between 1.03 - 1.10.

6.2.4 Deprotection of PtBS-b-PHEMATBDMS

Deprotection of PtBS-b-PHEMATBDMS into PtBS-b-PHEMA is shown in Figure 6.3 and was done using the following procedure. PtBS-b-PHEMATBDMS was placed into a small round bottom flask with a stir bar and dissolved in dichloromethane. Tertbutyl ammonium fluoride was added such that it was in two-molar excess compared to the amount of TBDMS in the BCP chain. The solution was then allowed to stir for 24 hours at room temperature. Next the solution was washed with deionized water three times, rotavaped, and then dried in a vacuum oven. In cases where dead PtBS homopolymer is present, the BCP is dissolved in methanol and then washed with hexane to extract out the majority of homopolymer.

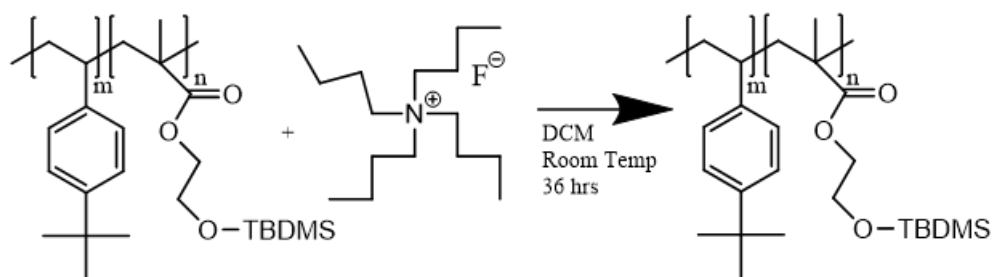


Figure 6.3: Reaction scheme for the deprotection of PtBS-b-PHEMATBDMS into PtBS-b-PHEMA.

6.2.5 Synthesis of Random Copolymer Underlayers

Figure 6.4 shows the two different underlayers were explored in the search for a neutral underlayer for thin films of PtBS-b-PHEMA to form perpendicular features on. For each random copolymer underlayer, 4-vinylbenzocyclobutene (BCB) was used as a thermal crosslinking agent. Each underlayer was polymerized via radical polymerization.

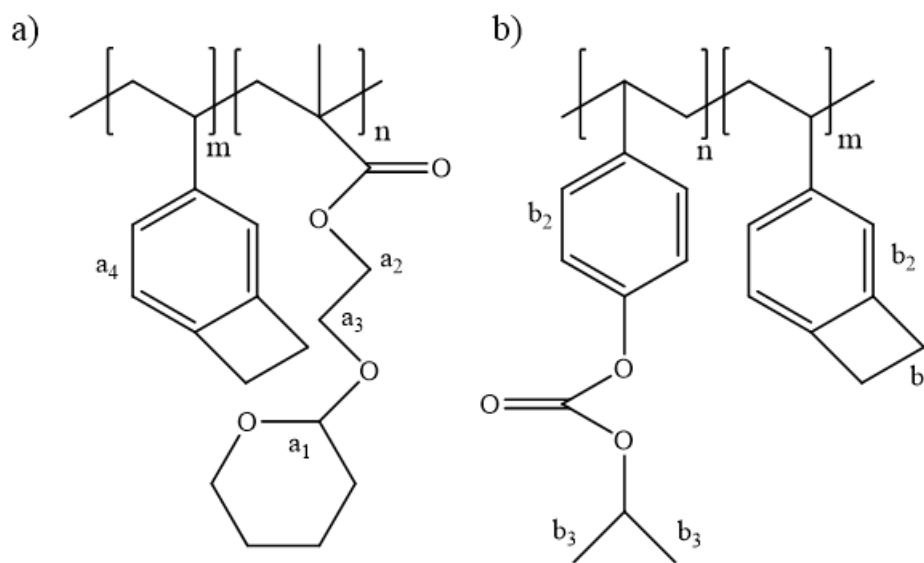


Figure 6.4: Structures of PHEMA-THP-r-BCB (a) and iPOC-r-BCB (b), the two underlayers explored in finding a neutral underlayer for PtBS-b-PHEMA.

The first underlayer explored was PHEMA-THP-r-BCB (Figure 6.4b). THP-protected HEMA was synthesized by reacting HEMA with a two-molar excess amount of 3,4-dihydro-

2H-pyran and 0.2 molar equivalent of trifluoroacetic acid in 5 mL of ethyl acetate at room temperature for 18 hours. The NMR of the protected HEMA-THP monomer is shown in Figure 6.5a where the alkyl protons (a_2 and a_3 , total of 4) of the HEMA are shown at 3.8 ppm (2) and 4.3 ppm (2) and the single proton between the oxygen of HEMA and the oxygen of THP (a_1) is found at a shift of 4.6 ppm. PHEMA-THP-r-BCB was synthesized by dissolving HEMA-THP, BCB, and AIBN in THF in a schlenk flask with a stir bar. The vessel was given three freeze-pump-thaw cycles, backfilling with nitrogen after each thaw. The solution was then placed in an oil bath at 75°C and stirred for 2 hours. Afterwards the solution was quenched, opened to the air, and precipitated in cold methanol. Figure 6.5b shows an example of the NMR for a typical PHEMA-THP-r-BCB underlayer with the alkyl protons (a_2 and a_3 , 4 total protons) at 3.8 ppm (2) and 4.3 ppm of PHEMA-THP and the phenyl protons of BCB at chemical shift of 6.5 - 6.9 ppm (a_4 , 3 protons total) are used to determine the relative amounts of PHEMA-THP and BCB in the random copolymer.

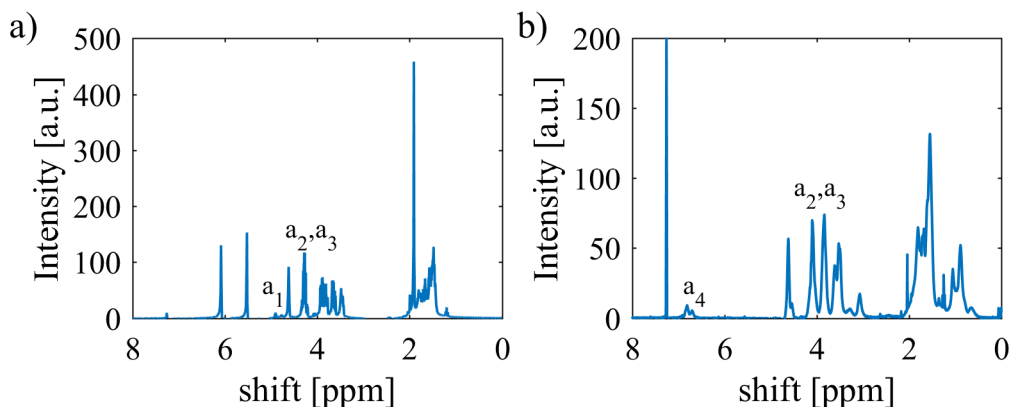


Figure 6.5: NMRs of the protected HEMA-THP monomer (a) and the random copolymer PHEMA-THP-r-BCB (b). Labels for PHEMA-THP-r-BCB correspond to protons at the positions labeled in Figure 6.4a.

The second underlayer explored was isopropylloxycarbonyl-r-BCB (iPOC-r-BCB, Figure 6.4b). Typical synthesis for iPOC-r-BCB is a three stage reaction. In the first step acetoxystyrene, BCB, and AIBN are dissolved in toluene, given three freeze-pump-thaw cycles (backfill with nitrogen) and then stirred at 75°C for 24 hours. The reaction was then

quenched, precipitated in cold hexane, and dried in a vacuum oven. NMR spectra identifies the acetoxystyrene by the acetyl protons (2.2 ppm, 3 protons) while the BCB identified by the butene protons (3.0 ppm, 4 protons). The product was then dissolved in dioxane, hydrazine hydrate was added slowly, dropwise, and allowed to stir for 12 hours to deprotect the poly(acetoxystyrene-r-BCB) into poly(hydroxystyrene-r-BCB). The product was then precipitated in deionized water and dried in a vacuum oven. NMR spectra of PHOST-r-BCB identifies the PHOST by the broad hydroxyl proton at 8 ppm (1 proton) and the BCB by its 4 protons at 3.0 ppm as previously mentioned. PHOST-r-BCB was then dissolved in ethyl acetate with triethylamine. Next iPOC-Cl was added dropwise to the solution and it was allowed to stir at room temperature for 24 hours. The product was then washed with 3% hydrochloric acid twice, poured over magnesium sulfate, and then precipitated in cold hexanes. Figure 6.6 shows an NMR of iPOC-r-BCB where the isopropyl protons of iPOC are shown at 1.4 ppm (c_3 , 6 protons) and the butene protons of BCB are shown at 3.0 ppm (b_2 , 4 protons).

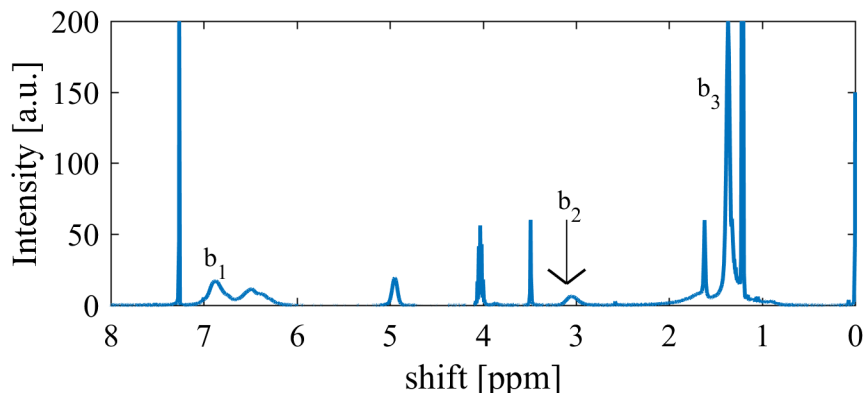


Figure 6.6: NMR for iPOC-r-BCB underlayer. Labels correspond to protons at the positions labeled in Figure 6.4b.

6.2.6 Characterization Methods

Characterization of the bulk BCP was done using GPC, NMR, IR, and SAXS. For NMR, while the protected PtBS-b-PHEMATBDMS was able to dissolve in deuterated chloro-

form, PtBS-b-PHEMA was not due to the high contrast in interactions between PtBS and PHEMA. Instead a mixture of chloroform and DMSO (75:25 by volume) was needed to dissolve PtBS-b-PHEMA. SAXS data was measured using the Xenocs Xeuss 2.0, Line Eraser using a Pilatus 300K SAXS detector. Bulk SAXS samples were annealed at 150°C for 12 hours prior to their measurement. Infrared spectroscopy was taken using a Thermo Nicolet IS10 under attenuated total reflectance mode using an MCT detector.

For thin film thicknesses were measured via ellipsometry. Underlayer preference was measured using the water contact angle of PtBS, PHEMA, and the underlayer. BCP thin films were annealed under vacuum at temperatures between 150°C and 210°C. Imaging of thin film morphology was done using a Zeiss Ultra60 FE-SEM and a Veeco ICON AFM with an AFM tip radius of 8 nm. Etch contrast between the blocks was done using a Harrick Plasma Cleaner. Samples were placed in a chamber, put under vacuum and O₂ plasma was generated from the air a radio frequency of 10.5W.

6.3 Results and Discussion

6.3.1 Synthesis

Figure 6.7 shows the structure of PtBS-b-PHEMA. The four phenyl protons of PtBS (a_1 in Figure 6.7) and the four alkyl protons of PHEMA (b_1 and c_1 of Figure 6.7) are used to determine the relative amounts of each block in the BCP. Since these three sets of protons do not alter due to the deprotection, their chemical shifts go relatively unchanged in the NMRs of PtBS-b-PHEMATBDMS and PtBS-b-PHEMA. Figure 6.8 and Figure 6.9 show the GPC traces and NMRs of PtBS-b-PHEMATBDMS and PtBS-b-PHEMA, respectively. The peaks for a_1 are found along the chemical shift of 6.2 - 7.3 ppm, while the shifts for b_1 and c_1 are found at 4.1 and 3.9 ppm, respectively. All PDIs of PtBS-b-PHEMA BCPs were measured as less than 1.13. The volume fraction of PHEMA in the BCP samples ranged from 0.28 - 0.52.

Infrared spectroscopy, under attenuated total reflection mode, of the protected and de-

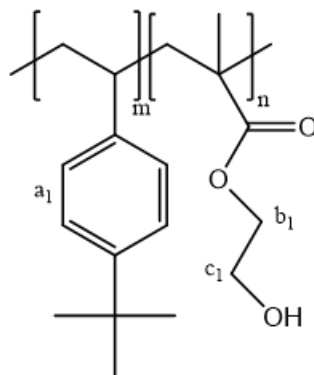


Figure 6.7: Structure of PtBS-b-PHEMA. The areas labeled the phenyl protons a_1 of PtBS and the alkyl protons b_1 and c_1 of PHEMA are used to characterize the relative volume fractions of the blocks.

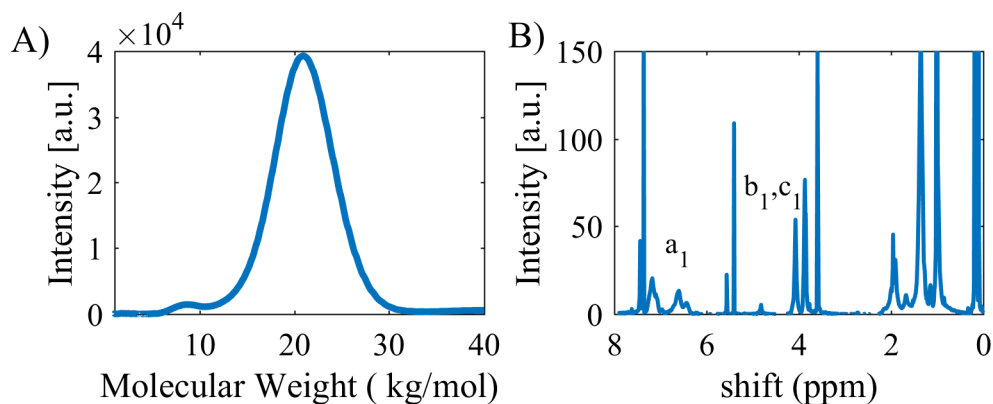


Figure 6.8: Examples of typical GPC traces (a) and NMRs (b) of PtBS-b-PHEMATBDMS

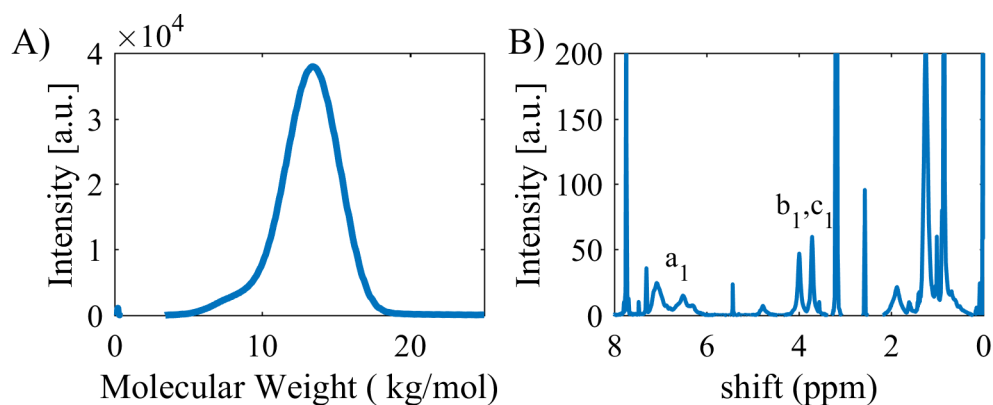


Figure 6.9: Examples of typical GPC traces (a) and NMRs (b) of the final product of PtBS-b-PHEMA.

protected BCP was done to ensure that a hydroxyl group had formed in the sample. Figure 6.10a shows the IR of PtBS-b-PHEMATBDMS with the absence of a hydroxyl peak and the appearance of silicon-carbon bonds, albeit in the fingerprint region of the scan. Figure 6.10b shows the IR of PtBS-b-PHEMA with a large peak at roughly 3500 cm^{-1} denoting the presence of hydroxyl groups in the BCP. There is also a disappearance of the silicon-carbon peaks in the fingerprint region as well. The combination of NMR and IR, and the GPC showing a relatively unchanged molecular weight distribution, is evidence that the synthesis of PtBS-b-PHEMA was a success.

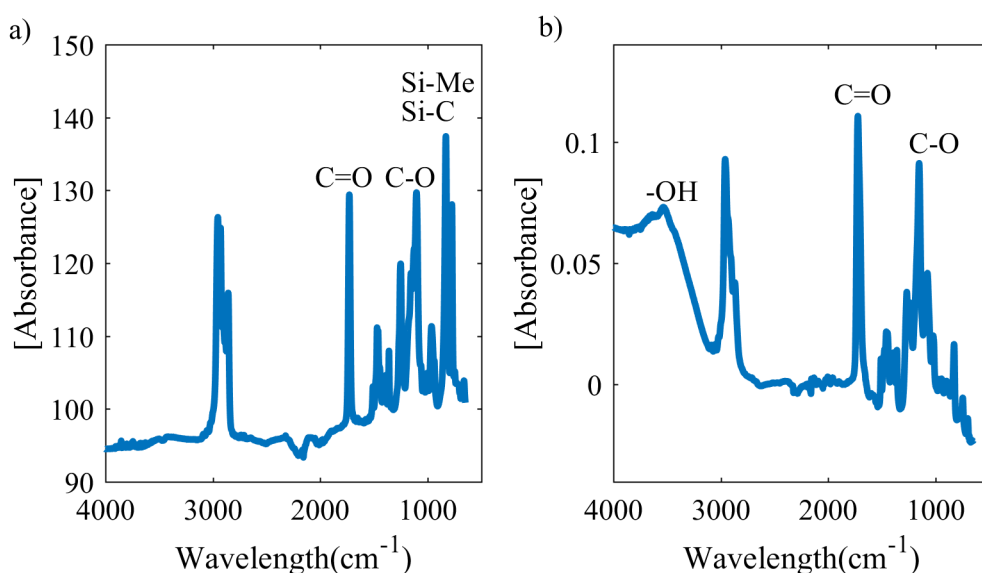


Figure 6.10: Infrared spectroscopy scans taken under attenuated total reflection mode for PtBS-b-PHEMATBDMS (a) and PtBS-b-PHEMA (b).

6.3.2 SAXS Characterization

SAXS measurements were taken of bulk BCP samples to determine the morphology and pitch of microphase separated features. Bulk samples were prepared by creating a mold for a disk with a 5 mm diameter and a 1 mm thickness. BCP powder was placed in the mold and dissolved with a few drops of solvent. The mold was then placed in a vacuum oven at 90°C for 30 minutes. The temperature was then raised to the desired annealing

temperature (150°C - 210°C) and allowed to anneal for 12 hours. The BCP disk was then peeled off the aluminum mold and SAXS was performed. Examples of the SAXS profiles of PtBS-b-PHEMA are shown in Figure 6.11.

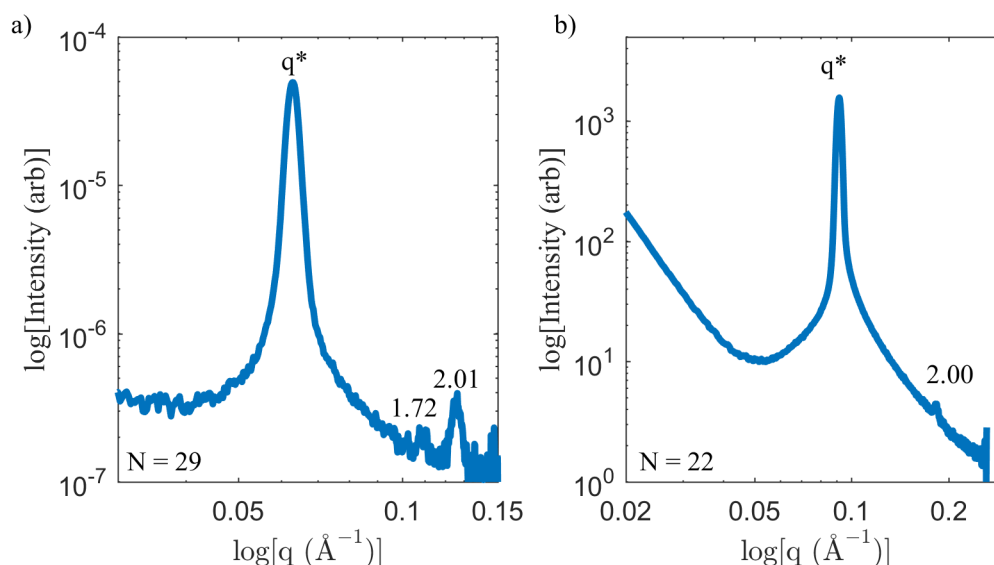


Figure 6.11: SAXS patterns of PtBS-b-PHEMA. The cylinder forming sample (a) has a $\phi_{PHEMA} = 0.36$, an $N = 29$, and was annealed at 150°C for 12 hours. The lamellae forming sample (b) has a $\phi_{PHEMA} = 0.52$, an $N = 22$, and was annealed at 210°C for 12 hours. The measured pitch by q^* for these samples are 10 nm (a) and 6.9 nm (b).

The position of the primary peak (q^*) of a scattering profile gives information on the pitch of the phase separated features, while the reflections of the q^* give information on the morphology. The majority of our samples showed a morphology of either hexagonally close packed cylinders (q^* reflections at $\sqrt{3}q^*$, $2q^*$, $\sqrt{7}q^*$, $3q^*$...) or lamellae (q^* reflections at $2q^*$, $3q^*$, $4q^*$...). [128] The more reflections present, the more ordered the features are. Figure 6.11a shows a cylinder forming sample ($\phi_{PHEMA} = 0.36$) with a pitch of 10 nm and reflections at $\sqrt{3}q^*$ and $2q^*$ while Figure 6.11b shows an example of a lamellae forming sample ($\phi_{PHEMA} = 0.52$) with a pitch 6.9 nm and a single reflection at $2q^*$. The sample in Figure 6.11b represents the lowest molecular weight sample produced with an $N = 22$. Despite this, the BCP was still able to phase separate, making it unable to determine this BCP's χ . However the fact that the PtBS-b-PHEMA was able to form features with a half-

pitch less than 4 nm is a clear indication of a high- χ material. A lower limit for estimate of the χ of PtBS-b-PHEMA, using the ODT as $\chi N = 10.5$, would be about 0.45.

Figure 6.12 shows a plot of the experimentally measured pitch of lamellae forming PtBS-b-PHEMA samples annealed at 150°C for 12 hours versus their degree of polymerization. A trendline was fit to this data (dashed line in Figure 6.12) to determine how the pitch scales with N . The exponent calculated by the trendline was 0.802. Given the weak reflection in Figure 6.11b of the $N = 22$ sample, it should be expected that $N = 22$ is in the proximity of the ODT. This would mean that the points shown in Figure 6.12 are expected to be in the intermediate segregation regime where the expected scaling of the pitch is $N^{0.8}$ - $N^{1.0}$. [118, 119] This matches well with the scaling calculated by the trendline in Figure 6.12.

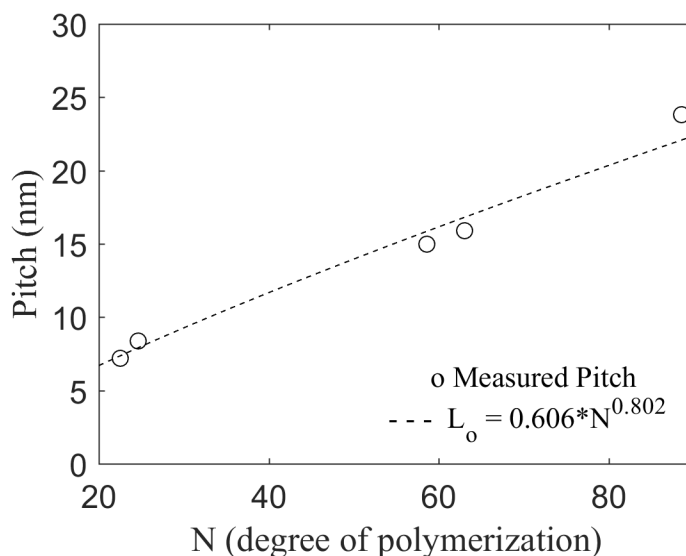


Figure 6.12: Plot of experimentally measured pitch (o) of lamellae forming PtBS-b-PHEMA at 150°C vs degree of polymerization. The dashed line is a trend line, with its equation in the bottom right of the plot.

6.3.3 Underlayer Characterization

The crosslinking procedure used was the same for all underlayers. After coating, the films were heated to 250°C for 10 minutes causing the BCB units to crosslink with one another. [129, 130] The water contact angle for PtBS and PHEMA was measured as 104° and 38°, respectively. A "neutral" underlayer here is described as one that provides perpendicular orientation to a lamellae forming BCP, and thus has no strong preference for either PtBS or PHEMA. First estimates would suggest a neutral underlayer would have a water contact angle near 71°, the average of PtBS and PHEMA's water contact angle. A random copolymer, PtBS-r-b-PHEMA-r-BCB, was originally synthesized but showed little variation in water contact angle with mole fraction of PHEMA in the BCP. Added, in order to get an underlayer with a different surface energy, a new random copolymer would need to be synthesized each time. The random copolymer PtBS-r-PHEMA-r-BCB was expected to be able to traverse the entire range of water contact angles. Given that this BCP is expected to be a high- χ material and will likely have a narrow window in contact angle, a faster method of varying the contact angle would be desirable compared to synthesis of a new underlayer for any change in contact angle.

PHEMA-THP-r-BCB was developed as an acid sensitive underlayer with the ability to change its surface energy. The random copolymer was dissolved in PGMEA with 15 wt% TPS-SbF₆ and filtered. The solution was spin coated onto a clean silicon wafer piece and the BCB units were crosslinked. Next the thin film was exposed to a specific dose of 248 nm wavelength light, and then baked at 100°C for two minutes. The exposure to 248 nm light causes an acid to form from the TPS-SbF₆ and the post exposure bake allows this acid to diffuse through the film and deprotect PHEMA-THP into PHEMA. [131] By varying the dose of light the film receives, the amount of deprotection can be controlled. After the post exposure bake, the film is then rinsed with PGMEA and baked at 90°C to remove any remaining solvent. Figure 6.13 shows the variance in water contact angle of PHEMA-THP-r-BCB with exposure dose. With no deprotection, PHEMA-THP-r-BCB

shows a water contact angle of about 66° . The random copolymer however is very sensitive to the acid concentration, dropping to around 55° after an exposure of only 5 mJ/cm^2 .

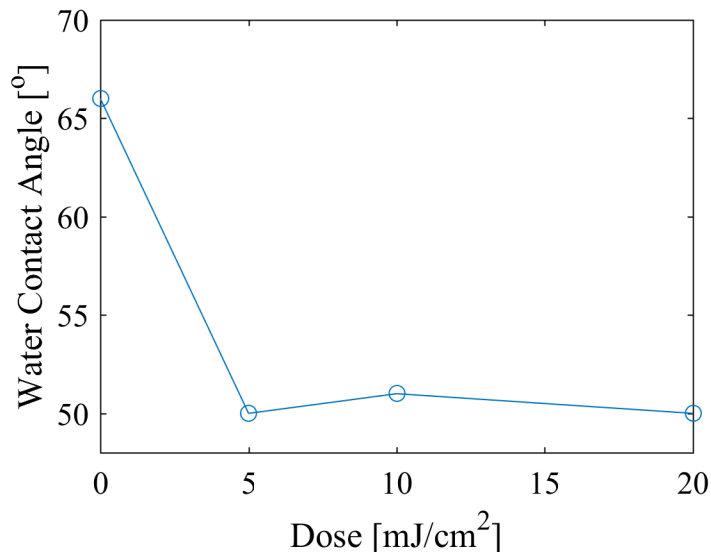


Figure 6.13: Water contact angle versus exposure of 248 nm DUV light to PHEMA-THP-r-BCB. Films were given a post exposure bake at 100°C for two minutes.

The random copolymer iPOC-r-BCB was developed similar to PHEMA-THP-r-BCB in that its surface energy too is sensitive to acid deprotection of the iPOC into PHOST. The iPOC-r-BCB was dissolved in PGMEA with 20wt% TPS-SbF₆, filtered, and then coated onto a clean wafer piece. After crosslinking the BCB, the film was exposed to 248 nm wavelength light and then given a post exposure bake at 170°C for two minutes. Figure 6.14 shows the water contact angle of iPOC-r-BCB is well controlled by the exposure dose and varies between 70° and 85° .

6.3.4 Thin Film BCP Self-Assembly

Thin films of PtBS-b-PHEMA were dissolved in THF, filtered, and coated onto silicon wafer pieces pre-coated with the crosslinked underlayers mentioned in the previous section. The thickness of the crosslinked underlayers used were typically in the range of 20 - 40 nm. To promote perpendicular orientation of the BCPs morphology, BCP thin films were coated

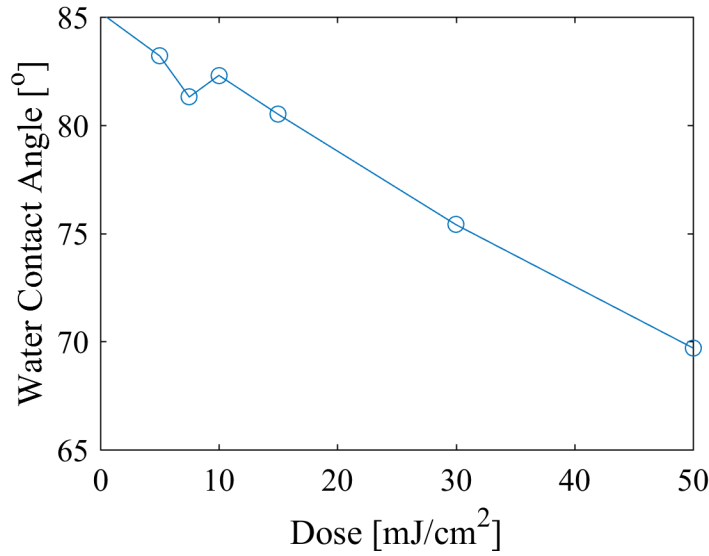


Figure 6.14: Water contact angle versus exposure of 248 nm light to iPOC-r-BCB. Films were given a post exposure bake at 170°C for two minutes.

at incommensurate thicknesses with their pitch. Thicknesses ranged between $1 \cdot L_o - 3 \cdot L_o$. For each underlayer, a lamellae forming and a PHEMA-cylinder forming BCP sample was coated on a silicon wafer piece. While lamellae forming BCPs prefer a neutral underlayer in order to orient perpendicular to the substrate, PHEMA-cylinder forming BCPs would prefer an underlayer slightly preferential to PHEMA in order for the PHEMA-cylinders to orient perpendicular. The three BCP samples used on each underlayer was a lamellae forming BCP (L1) with a pitch of 15 nm ($\phi_{PHEMA} = 0.45$) and an $N = 58$, a cylinder-forming BCP (C1) with a 10 nm pitch ($\phi_{PHEMA} = 0.36$) and an $N = 29$, and a suggested perforated lamellae forming BCP (PL1) with a 24 nm pitch ($\phi_{PHEMA} = 0.34$) and an $N = 88$. While the SAXS for PL1 at first glance appears to have a lamellar morphology, after thin film annealing of the sample, perpendicularly oriented cylindrical features were observed. Due to the conflicting results from SAXS and thin film samples, it is suggested that this sample is actually forming a metastable perforated lamellae morphology. These samples were chosen due to them having the largest pitches for their morphology making them the easiest to focus on in the SEM. After the BCP film was coated onto the crosslinked

underlayer, the sample was baked at 90°C for two minutes to remove the casting solvent. The BCP thin film was then placed under vacuum (< 25 torr) and the temperature was increased to 160°C and the films were allowed to anneal for 12 hours.

Table 6.1 gives an overview of the thin film results as examined by SEM where the symbol to the right of data point represents the orientation of the features as parallel or perpendicular. Depending on the underlayer preference, islands and holes may form when the film thickness is incommensurate with the BCP's pitch. [132] For a neutral underlayer, these islands and holes will not form at incommensurate film thicknesses. These islands and holes are visible by the SEM. Topography was seen for all C1 samples with featureless SEM scans when zoomed in and focused. This suggests either that the windows for perpendicularly oriented features is either very narrow or that a more PHEMA-preferential (the minority block of the cylinder forming BCP) underlayer is required. Figure 6.15 shows the peak error force of an AFM scan of C1 on an underlayer with a 75° water contact angle. The peak error force is a measure of the overshoot by the AFM tip when it approaches a region with topography. Cylinders are oriented parallel to the substrate giving the appearance of striped patterns over the scan. The peak error force shows these stripes implying that the buried cylinders are causing a slight topography in the underlayer. The inset of Figure 6.15 shows the 2D-fast fourier transform (2D-FFT) of the striped pattern and a profile of the peak error force along several parallel cylinders is shown just below the AFM scan. Both the 2D-FFT and the profile along some select cylinders show an average pitch of about 12 nm, in close agreement with the bulk pitch of 10 nm determined by SAXS.

Table 6.1: Summary of orientation of BCP sample with different morphologies on thin films of various water contact angles.

Underlayer Contact Angle:	50°	66°	75°	80°
L1 (lamellae)				
PL1 (perforated lamellae)			$\perp c$	
C1 (PHEMA cylinders)				

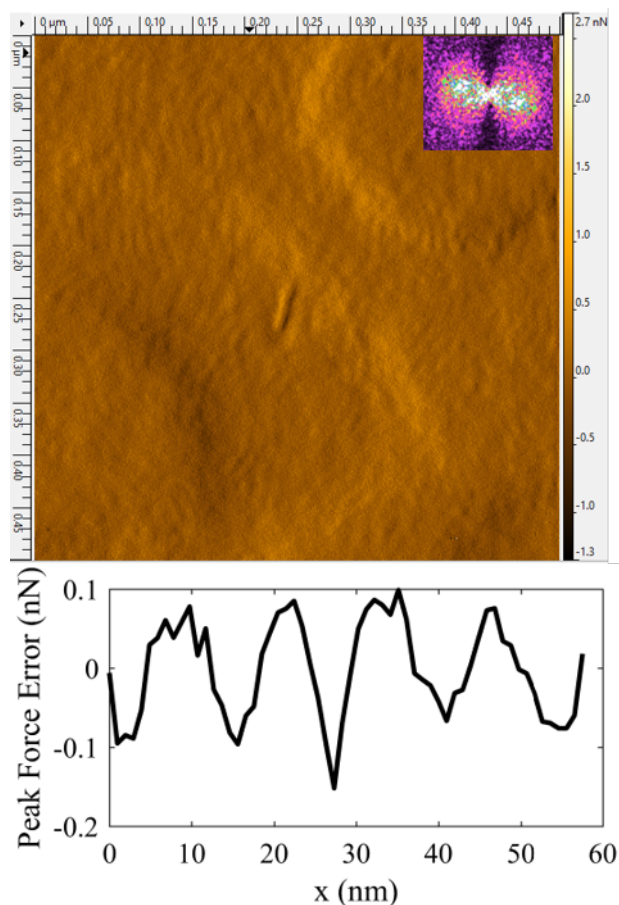


Figure 6.15: The peak force error of an AFM scan over a the cylinder forming C1 sample. The inset shows a 2D-FFT of the parallel oriented cylinders. Just below the AFM scan is a profile of the peak force error over a few select cylinders. Both the profile and the 2D-FFT give pitch estimates of about 12 nm which is in good agreement with the SAXS determined pitch of 10 nm.

Thin film samples of L1 were featureless in the SEM and AFM. Parallel oriented lamellae stack in layers, and would appear featureless from top down scans of either imaging technique. No large scale topography of islands and holes though were seen for L1 samples on underlayers shown in Table 6.1. The perforated lamellae sample (PL1) showed some island/hole topography only while on a 50° water contact angle underlayer. For perforated lamellae the only instance when features should not be present at the surface is when the thickness is such that the perforating block's lamellae is at the surface. SEMs for PL1 thin films atop 66° and 80° water contact angle underlayers displayed a mixture

of feature and featureless regions at the surface which may be explained by variations in local film thickness. At a 75° contact angle however perpendicular cylindrical features ($\perp c$ in Table 6.1) were seen as shown in Figure 6.16. Due to the presence of these cylinders conflicting with the lamellar morphology SAXS profile, this sample is suggested to be perforated lamellae. The 2D-FFT inset measure an average pitch between perforations of about 26 nm, in good agreement with the bulk pitch measurement of 24 nm by SAXS. The presence of these cylinders means that neither the PtBS nor the PHEMA block prefers to be at the free surface so greatly that a wetting layer inherently forms there. While cylinder and lamellae forming samples have yet to show perpendicular oriented features, the fact that no wetting layer should form at the free surface means that these feature's orientation can be controlled by underlayer preference.

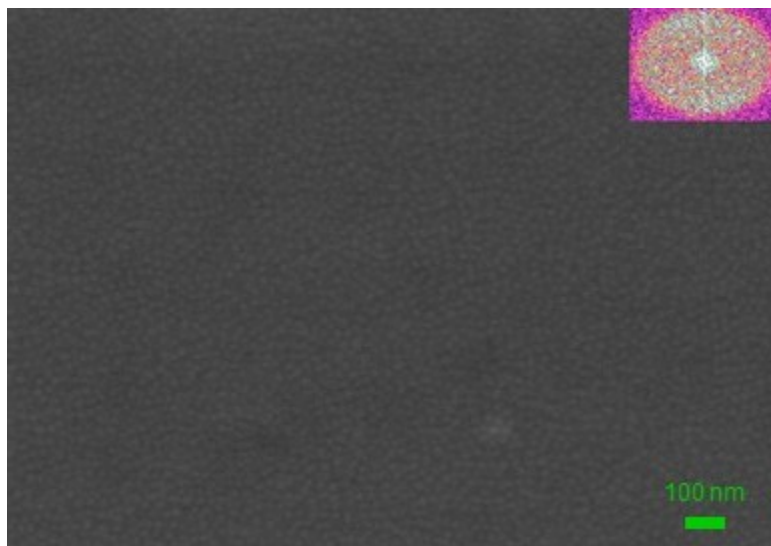


Figure 6.16: SEM of perforated lamellae sample (PL1) on a 75° water contact angle underlayer. The inset 2D-FFT measures the average pitch at 26 nm, in close agreement with the SAXS determined pitch of 24 nm for this sample.

6.3.5 Etch Contrast

To be useful as a lithographic mask, BCPs must be composed of blocks with an etch contrast, meaning one block needs to be able to be etched faster than the other. To determine

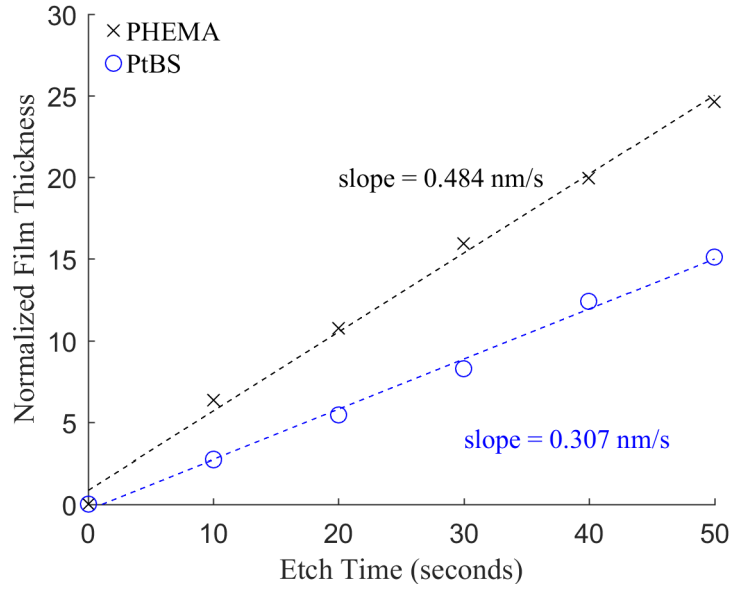


Figure 6.17: SEM of perforated lamellae sample (PL1) on a 75° water contact angle underlayer. The inset 2D-FFT measures the average pitch at 26 nm, in close agreement with the SAXS determined pitch of 24 nm for this sample.

the etch contrast for PtBS-b-PHEMA, an etch study on thin films of homopolymer PtBS and PHEMA was conducted. While the O_2 plasma used in this etch study will not be used to etch silicon, it is useful as a proxy for determining etch contrast between PtBS and PHEMA. The Ohnishi parameter (O.N.) can be used to determine the expected etch contrast between two polymers based on the ratios of carbon atoms in each polymer's repeat unit.[133, 134, 135] Equation 6.1 shows the Ohnishi parameter (O.N.) is calculated as the ratio of the total number of atoms (N_{total}) in a repeat unit to the difference in the number of carbons (N_C) and oxygen (N_O) atoms.

$$O.N. = \frac{N_{total}}{N_C - N_O} \quad (6.1)$$

For PtBS $O.N._{PtBS} = 2.3$ while for $O.N._{PHEMA} = 6.3$. For comparison, $O.N._{PPMA} = 4.2$, and PtBS and PPMA had an etch contrast of roughly 2 (very near to the 2.3 etch contrast found in Chapter 5 for PtBS-b-PPMA). This means that PHEMA is expected to

etch roughly three times faster than PtBS.

Homopolymer PtBS and PHEMA were dissolved in PGMEA and DMF, respectively, and coated on clean silicon wafer pieces. One at a time, the films were placed into an O₂ plasma etcher, put under vacuum, and then etched (RF: 10.5W) in 10 second intervals. Figure 6.17 shows the amount etched from the two films at each time interval with the slopes representing the etch rate. PHEMA homopolymer was found to etch 60% faster than PtBS under these conditions. It should be noted that the etcher used for this study is not nearly as well controlled as the etcher used for PtBS-b-PPMA in the previous chapter. The conditions inside the etcher may need to be optimized as well with a proper concentration of O₂ plasma rather than just using O₂ from the air that leaks into the system.

However even if PHEMA did have a low etch contrast with PtBS, an alternative route does exist to increase that contrast. Due to PHEMA have a native hydroxyl group, sequential infiltration synthesis could be used to grow layers of titanium or aluminum oxides in the PHEMA region, increasing the etch contrast between the two BCPs. [136]

6.4 Conclusions

PtBS-b-PHEMA, a new high- χ BCP, was synthesized by anionic polymerization and ATRP with low PDIs (< 1.15). Bulk SAXS profiles of PtBS-b-PHEMA show that the BCP can form lamellae with a sub-7 nm pitch and cylinders with a 10 nm pitch. Radical polymerized random copolymer underlayers with the ability to change their surface energy via acid-catalyzed reactions were used for thin film studies. PHEMA-THP-r-BCB and iPOC-r-BCB were able to successfully modulated their surface energies over a water contact angle range of 50°-66° and 70°-85°, respectively. Perpendicular orientation of features was only found for a perforated lamellae sample implying that a top coat may not be required for the BCP. Further thin film studies are need to find an underlayer that will perpendicularly oriented features to the substrate for lamellae- and cylinder- forming samples of PtBS-b-PHEMA. Due to the high χ of PtBS-b-PHEMA, a narrow window in surface energies may exist

for these to occur. An etch study was conducted for PtBS and PHEMA homopolymers showing that PHEMA etches about 60% faster than PtBS under O₂ plasma. This etch contrast is expected to increase by using a more well controlled etcher.

CHAPTER 7

CONCLUSIONS & RECOMMENDATIONS

7.1 Conclusions

Block copolymer directed self-assembly rivals EUV lithography for next generation extensions to optical lithography. While significant attention and funds are being placed on implementing EUV lithography, block copolymers are not being tossed aside. EUV tools are very expensive and not every company will be able to afford one, let alone a fleet, of the tools for chip manufacturing. Larger companies, such as Intel, may be able to afford experimenting with EUV, but the majority of smaller companies still see an interest in furthering the research of DSA-BCPs. The major obstacles standing in the way of BCP implementation are still synthesis of BCPs able to reach small feature sizes, defectivity, and line edge roughness/line width roughness. This work takes a deeper look into how the underlayer can affect the likelihood of defects occurring using simulations. In addition, simulations were used to probe how LER and LWR vary with the addition of a common blend component of BCPs, homopolymer. Lastly, the library of high and low χ BCP materials were extended with PtBS-b-PHEMA and PtBS-b-PPMA, respectively.

In Chapter 3 coarse grained molecular dynamics simulations were used to probe the effects of defect order and underlayer patterning properties on the relative free energy of defects in BCP thin films on chemoepitaxially patterned underlayers. It was found that for low defect orders (1 - 3) relative defect free energy increased linearly with increased spacing between the cores of the defect. However at a defect order of 4 and 5, relative free energy decreased with respect to that at defect order 3, meaning the defect was more likely to occur. This was assumed to be due to a release of strain in the defect's lamellae caused by the cores of the defect being too close to one another. The relative free energy

of defects were also shown to decrease with increasing density multiplication, showing the trade offs caused by the periodicity of the pinning stripe. Pinning stripe position relative to the defect was also varied, determining that the dislocation's size is most unstable when a pinning stripe is just to the interior of its terminating block. Ultimately, despite all of these variations, relative defect free energies were still found to be in the 100s of $k_B T$. This further bolsters the suspicion that defects are highly thermodynamically unfavorable, and that the high defectivity levels observed experimentally are likely due to the defects being kinetically trapped.

Chapter 4 probed the effect of homopolymer concentration on LER and LWR for BCP thin films using the same coarse grained molecular dynamics simulations. LER and LWR both increased with homopolymer concentration, however; at low homopolymer concentration (10% or less) the top and bottom of the films began to experience significantly different LER and LWR. In addition, at very high concentrations of homopolymer (26% or greater), LER in pinned and unpinned lamellae began to show a dependence on pinning stripe width. Differing LER and LWR between the top and bottom of the film is likely due to homopolymer segregation and the composition of the background region. For instance, at the pinning stripe, homopolymer almost exclusively wet the substrate, with very little being seen at the free surface. The degree of homopolymer segregation varies depending on the lamellae in question and the background region composition, with lamellae far from the pinning stripe more likely to have a uniform composition of homopolymer through the depth of the film for a more neutral background region. The source of increased LER and LWR with homopolymer concentration however appears to be the fluctuations in homopolymer concentration along the length of the lamellae. Depending on the concentration along this length, lamellae interfaces are placed at different positions and the interface becomes more uncorrelated with its neighbors. While discrepancies between LER and LWR at the top and bottom of a film can be detrimental for pattern transfer, the effect may only be significant for high homopolymer concentrations. This means that the tunability of BCP

pitch via homopolymer blending is limited

A new low χ BCP material, PtBS-b-PPMA, was synthesized via anionic polymerization in Chapter 5. Bulk small angle X-ray scattering was used along with Leibler Theory to calculate a $\chi = 0.027$, roughly 67% the χ of PS-b-PMMA. The decrease in χ is suspected to be due to the increase in hydrophobicity of the two blocks compared to PS and PMMA. PtBS-b-PPMA was found to produce a minimum lamellar pitch of about 40 nm with a maximum lamellar pitch produced in this work of being 83 nm. A random copolymer was synthesized via radical polymerization using PtBS, PPMA, and BCB as a crosslinking agent to produce a neutral underlayer for PtBS-b-PPMA. PPMA was determined to be a thermally sensitive block via thermogravimetric analysis, limiting the BCP to thermal annealing at temperatures under 180°C. Given the glass transition temperatures of the two blocks, lengthy annealing times were needed for the BCP to form a fingerprint lamellae pattern. An etch study revealed that there does exist an etch contrast between the two blocks, with PPMA etching 2.3 times faster than PtBS. Low χ materials are suspected to increase defect annihilation kinetics compared to their high χ counterparts which may lead to high N , low χ BCPs finding applications as photonic crystals.

In Chapter 6 a new high χ material, PtBS-b-PHEMA, was synthesized via anionic and atom-transfer radical polymerization. While the χ of the BCP is not currently known, SAXS results show that PtBS-b-PHEMA has the capability to phase separate into sub-7 nm pitch line-space patterns. This leads to the conclusion that the χ of PtBS-b-PHEMA must be significantly higher than PS-b-PMMA whose limit is near 18 nm pitch features. The cause for the increase in χ is likely due to the increase in contrast between the hydrophobic block PtBS and the hydrophilic block PHEMA. However with an increase in χ comes further difficulty in finding a neutral underlayer for thin film BCP studies. Two new crosslink-able underlayers were developed with the ability to tune their surface chemistry in the hopes of finding a neutral underlayer for the BCP. While no neutral underlayer was found to produce perpendicular lamellae, one was found to produce perpendicularly ori-

ented features for a perforated lamellae sample. This means that at least a wetting layer of PtBS or PHEMA does not inherently form at the free surface, and that perhaps the window for a neutral underlayer is only very narrow.

7.2 Future Work & Recommendations

Specifically for simulation work there are several directions to continue the research. It would be interesting to determine the effect that homopolymer concentration and PDI have on the likelihood of defect formation. For homopolymer concentration it's possible this will be complicated by concentration fluctuations along the lamellae length. While for PDI it would be more of an issue in classifying the type of PDI for the BCP. For instance, PDI is typically thought of as the variation in N of the BCP, so by one measure the PDI could increase by having 50/50 A/B block copolymer chains with a distribution of their total number of beads (N). However, there could also be a distribution in the individual chain's relative fraction of A-beads and B-beads with or without changing the total N of the chains with respect to the N of the BCP.

In addition the effect of χ , N , and χN on defect free energy and LER/LWR for these blended systems would also be valuable information. For homopolymer/BCP blend simulations, how N of the homopolymer in relation to the BCP affects the process window could also be an interesting route to explore. The major limitations for the previously mentioned work would be simulation time. Especially for experiments where N is changed of the BCP, larger simulations would need be run meaning more beads and longer computer run times. For thermodynamic integration in particular, turning on the external potential for an $8L_o \times 6L_o \times L_o$ sized simulations (where $L_o = 11.8631$ nm) takes roughly 10 days. Fortunately these run times can be lessened by an upgrade from the current 580s Nvidia graphics cards being used to something closer to a 980 or 1080 Nvidia graphics card. However benchmarks will have to be run to determine exactly what the boost in simulation speed is.

With regards to PtBS-b-PPMA and PtBS-b-PHEMA, the synthesis procedure could use a small improvement. The current procedure appears to produce about a 10% contamination of homopolymer due to impurities. It is suspected this could be minimized by further purification of the monomers, specifically DPE or the methacrylate-based monomer. For PtBS-b-PPMA, further research into optimal thin film annealing conditions would be useful. Either higher temperatures with shorter annealing times are necessary or further exploration into finding a neutral solvent for the solvent annealing of the BCP are needed. It is recommended that for finding a neutral solvent a flow system be built where two streams mix with one another before coming in contact with the film. One stream should be an inert gas and one stream should be an inert gas bubbled through a particular solvent. This will allow the user to control solvent composition easily in the gas mixture that interacts with the thin film. This procedure should be done for films of the individual homopolymers of each block of the BCP. Once a proper annealing condition is found for PtBS-b-PPMA, further research can be conducted in its DSA.

For PtBS-b-PHEMA's synthesis, further investigation into the deprotection of PtBS-b-PHEMATBDMS to PtBS-b-PHEMA should be conducted. The deprotection reaction was not always consistent, sometimes leading to broad, multi-peaked traces in the GPC. The cause for this is currently unknown. A neutral underlayer needs to be found for PtBS-b-PHEMA as well. This could be done by performing further thin film experiments on underlayers with contact angles between 38° and 80° that were not studied in this work. Rather than using an SEM for such experiments (which requires difficult focusing), an AFM is suggested to be used which can offer higher certainty in the determination of features at the surface of the film. Once a neutral underlayer is found, various annealing temperatures can be studied to find an optimal annealing condition for the BCP.

Appendices

APPENDIX A

LEIBLER THEORY

Leibler characterized block copolymers (BCPs) using small-angle x-ray scattering (SAXS).[26] Using his equations, for a BCP below the order-disorder transition (ODT), χ can be calculated. BCPs were modeled as taking a gaussian, ideal shape, hence the need for the BCP to be below the ODT where chains are expected to conform to this. For convenience, a term x is made and described by Equation A.1.

$$x = \frac{q^2 \cdot N \cdot a_{BCP}^2}{6} \quad (\text{A.1})$$

Here, q is the scattering vector, N is the degree of polymerization and a_{BCP} is the statistical segment length of the BCP. The scattering intensity of the BCP chains are described by the debye function $g_1(f, x)$ in Equation A.2.

$$g_1(f, x) = \frac{2 \cdot [f \cdot x + \exp(-f \cdot x) - 1]}{x^2} \quad (\text{A.2})$$

In Equation A.2, f is the volume fraction of a particular block in the BCP. The debye function is calculated for both f and $1 - f$, and then scattering interaction between these two blocks is described by $F(x)$.

$$F(x) = \frac{g_1(1, x)}{g_1(f, x) \cdot g_1(1 - f, x) - 0.25 \cdot [g_1(1, x) - g_1(f, x) - g_1(1 - f, x)]^2} \quad (\text{A.3})$$

Finally, the totally scattering intensity ($S(q)$) is described by Equation A.4.

$$S(q) = \frac{N}{F(x) - 2 \cdot \chi \cdot N} \quad (\text{A.4})$$

In determining χ for a BCP, a SAXS measurement must first be taken for a sample below the ODT. A BCP can be determined as below the ODT by a broadening of the primary peak in the scattering profile. Then, the primary peak of the scattering profile is fit to Equation A.4 with χ and a_{BCP} as fitting parameters. A good check for this procedure is to compare the fitted a_{BCP} to literature values of a for each block, with a_{BCP} expected to be an average of the two. A proportionality constant K may be necessary to multiply $S(q)$ by in order to fit Equation ?? to real scattering data.

APPENDIX B

EXTERNAL POTENTIALS

The following sets of matrices are the table potentials used to create the defects in Chapter 3 of varying widths. The outside area not affected by these tables potentials, and defect-free simulations, felt the external potential described by Equation 1.2. For each Defect Order below, FX and FY are the forces felts by beads in a particular area in the x- and y-direction, respectively. The value of V is the potential that the beads feel in that area.

B.0.1 Defect Order = 1

if($m_{index} == 1 \&\& m_{pos.x} > (-2 * pitch) \&\& m_{pos.x} < (2 * pitch) \&\& m_{pos.y} > (-2.0 * pitch) \&\& m_{pos.y} < (2.0 * pitch)$)

FX[0][0] = 0.2359; FX[1][0] = 3.0114; FX[2][0] = 9.0787; FX[3][0] = 9.3974; FX[4][0] = 3.0958; FX[5][0] = 0.2351; FX[6][0] = -0.0243; FX[7][0] = -1.6141; FX[8][0] = -7.2123; FX[9][0] = -9.9438; FX[10][0] = -5.1749; FX[11][0] = -0.8967; FX[12][0] = 0.0071; FX[13][0] = 0.9680; FX[14][0] = 4.5663; FX[15][0] = 9.5656; FX[16][0] = 7.5419; FX[17][0] = 2.1186; FX[18][0] = 0.1177; FX[19][0] = -0.3142; FX[20][0] = -3.4145; FX[21][0] = -9.1589; FX[22][0] = -8.8042; FX[23][0] = -3.0355; FX[24][0] = -0.3265; FX[25][0] = 0.1841; FX[0][1] = 0.2287; FX[1][1] = 2.9244; FX[2][1] = 9.0400; FX[3][1] = 9.5497; FX[4][1] = 3.1250; FX[5][1] = 0.1687; FX[6][1] = -0.0137; FX[7][1] = -1.3474; FX[8][1] = -7.0752; FX[9][1] = -10.1802; FX[10][1] = -5.4279; FX[11][1] = -0.9291; FX[12][1] = -0.0025; FX[13][1] = 0.9378; FX[14][1] = 4.7841; FX[15][1] = 10.1246; FX[16][1] = 7.3366; FX[17][1] = 1.7468; FX[18][1] = 0.0511; FX[19][1] = -0.1817; FX[20][1] = -3.2442; FX[21][1] = -9.2095; FX[22][1] = -8.7585; FX[23][1] = -3.1798; FX[24][1] = -0.4769; FX[25][1] = 0.1039; FX[0][2] = 0.1421; FX[1][2] = 2.7512; FX[2][2] = 8.7182; FX[3][2] = 9.5917; FX[4][2] = 3.5049; FX[5][2] = 0.2489; FX[6][2] = -0.0183; FX[7][2]

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0.0060; FY[20][50] = -0.2177; FY[21][50] = -0.3383; FY[22][50] = -0.1813; FY[23][50] = 0.0065; FY[24][50] = -0.0080; FY[25][50] = -0.0017; FY[0][51] = -0.0124; FY[1][51] = 0.0273; FY[2][51] = 0.2647; FY[3][51] = 0.1931; FY[4][51] = -0.1096; FY[5][51] = -0.1132; FY[6][51] = -0.0171; FY[7][51] = -0.0041; FY[8][51] = -0.1754; FY[9][51] = -0.6207; FY[10][51] = -0.2949; FY[11][51] = -0.0497; FY[12][51] = -0.0398; FY[13][51] = -0.1172; FY[14][51] = -0.2072; FY[15][51] = -0.3744; FY[16][51] = -0.5135; FY[17][51] = -0.0412; FY[18][51] = -0.0216; FY[19][51] = -0.0918; FY[20][51] = -0.0671; FY[21][51] = -0.3529; FY[22][51] = -0.5704; FY[23][51] = -0.1126; FY[24][51] = 0.0077; FY[25][51] = 0.0054; V[0][0] = 0.9996; V[1][0] = 0.9038; V[2][0] = 0.4177; V[3][0] = -0.3952; V[4][0] = -0.9012; V[5][0] = -0.9968; V[6][0] = -1.0026; V[7][0] = -0.9622; V[8][0] = -0.6292; V[9][0] = 0.1174; V[10][0] = 0.7557; V[11][0] = 0.9673; V[12][0] = 0.9904; V[13][0] = 0.9612; V[14][0] = 0.7711; V[15][0] = 0.1780; V[16][0] = -0.5627; V[17][0] = -0.9402; V[18][0] = -1.0011; V[19][0] = -0.9982; V[20][0] = -0.8827; V[21][0] = -0.3691; V[22][0] = 0.4147; V[23][0] = 0.8927; V[24][0] = 0.9944; V[25][0] = 0.9961; V[0][1] = 0.9967; V[1][1] = 0.9039; V[2][1] = 0.4263; V[3][1] = -0.3935; V[4][1] = -0.9079; V[5][1] = -1.0009; V[6][1] = -1.0032; V[7][1] = -0.9731; V[8][1] = -0.6611; V[9][1] = 0.0893; V[10][1] = 0.7531; V[11][1] = 0.9733; V[12][1] = 0.9989; V[13][1] = 0.9693; V[14][1] = 0.7784; V[15][1] = 0.1450; V[16][1] = -0.6108; V[17][1] = -0.9586; V[18][1] = -1.0024; V[19][1] = -1.0015; V[20][1] = -0.9036; V[21][1] = -0.3895; V[22][1] = 0.3910; V[23][1] = 0.8733; V[24][1] = 0.9891; V[25][1] = 0.9985; V[0][2] = 0.9926; V[1][2] = 0.9106; V[2][2] = 0.4518; V[3][2] = -0.3498; V[4][2] = -0.8878; V[5][2] = -1.0003; V[6][2] = -1.0041; V[7][2] = -0.9683; V[8][2] = -0.6496; V[9][2] = 0.1069; V[10][2] = 0.7762; V[11][2] = 0.9868; V[12][2] = 1.0020; V[13][2] = 0.9745; V[14][2] = 0.7795; V[15][2] = 0.1303; V[16][2] = -0.6424; V[17][2] = -0.9679; V[18][2] = -1.0006; V[19][2] = -1.0022; V[20][2] = -0.9033; V[21][2] = -0.3918; V[22][2] = 0.3863; V[23][2] = 0.8670; V[24][2] = 0.9873; V[25][2] = 0.9975; V[0][3] = 0.9907; V[1][3] = 0.9129; V[2][3] = 0.4830; V[3][3] = -0.2974; V[4][3] = -0.8564; V[5][3] = -0.9935; V[6][3] =

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 $V[10][7] = 0.8232$; $V[11][7] = 0.9784$; $V[12][7] = 0.9781$; $V[13][7] = 0.9304$; $V[14][7] =$
 0.6741 ; $V[15][7] = -0.0047$; $V[16][7] = -0.6921$; $V[17][7] = -0.9637$; $V[18][7] = -1.0011$;
 $V[19][7] = -0.9980$; $V[20][7] = -0.8606$; $V[21][7] = -0.2572$; $V[22][7] = 0.5539$; $V[23][7]$

= 0.9542; $V[24][7] = 1.0013$; $V[25][7] = 0.9948$; $V[0][8] = 1.0034$; $V[1][8] = 0.9435$;
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 -1.0044 ; $V[7][8] = -0.9307$; $V[8][8] = -0.4961$; $V[9][8] = 0.2804$; $V[10][8] = 0.8395$;
 $V[11][8] = 0.9901$; $V[12][8] = 0.9871$; $V[13][8] = 0.9330$; $V[14][8] = 0.6289$; $V[15][8] =$
 -0.0814 ; $V[16][8] = -0.7447$; $V[17][8] = -0.9793$; $V[18][8] = -1.0025$; $V[19][8] = -0.9959$;
 $V[20][8] = -0.8467$; $V[21][8] = -0.2120$; $V[22][8] = 0.5920$; $V[23][8] = 0.9642$; $V[24][8]$
 $= 1.0036$; $V[25][8] = 0.9944$; $V[0][9] = 0.9990$; $V[1][9] = 0.9121$; $V[2][9] = 0.4308$;
 $V[3][9] = -0.3750$; $V[4][9] = -0.8855$; $V[5][9] = -0.9967$; $V[6][9] = -1.0051$; $V[7][9] =$
 -0.9303 ; $V[8][9] = -0.5005$; $V[9][9] = 0.2654$; $V[10][9] = 0.8307$; $V[11][9] = 0.9873$;
 $V[12][9] = 0.9951$; $V[13][9] = 0.9327$; $V[14][9] = 0.6244$; $V[15][9] = -0.0972$; $V[16][9] =$
 -0.7597 ; $V[17][9] = -0.9887$; $V[18][9] = -1.0025$; $V[19][9] = -0.9980$; $V[20][9] = -0.8236$;
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 $= 0.9971$; $V[0][10] = 0.9975$; $V[1][10] = 0.9080$; $V[2][10] = 0.4071$; $V[3][10] = -0.3866$;
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 $= -0.4920$; $V[9][10] = 0.2747$; $V[10][10] = 0.8177$; $V[11][10] = 0.9787$; $V[12][10] =$
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 0.9696 ; $V[12][11] = 0.9856$; $V[13][11] = 0.9217$; $V[14][11] = 0.6099$; $V[15][11] = -$
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 0.9687 ; $V[24][11] = 1.0009$; $V[25][11] = 0.9917$; $V[0][12] = 0.9992$; $V[1][12] = 0.9368$;
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0.8358; $V[11][12] = 0.9629$; $V[12][12] = 0.9771$; $V[13][12] = 0.9139$; $V[14][12] = 0.5645$;
 $V[15][12] = -0.1409$; $V[16][12] = -0.7538$; $V[17][12] = -0.9803$; $V[18][12] = -1.0012$;
 $V[19][12] = -0.9819$; $V[20][12] = -0.7556$; $V[21][12] = -0.0707$; $V[22][12] = 0.6738$;
 $V[23][12] = 0.9648$; $V[24][12] = 1.0016$; $V[25][12] = 0.9846$; $V[0][13] = 0.9994$; $V[1][13]$
 $= 0.9246$; $V[2][13] = 0.4604$; $V[3][13] = -0.3574$; $V[4][13] = -0.8854$; $V[5][13] = -0.9988$;
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 $= 0.8681$; $V[11][13] = 0.9644$; $V[12][13] = 0.9702$; $V[13][13] = 0.8909$; $V[14][13] =$
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 $= -1.0009$; $V[6][14] = -1.0031$; $V[7][14] = -0.8845$; $V[8][14] = -0.3717$; $V[9][14] = 0.4124$;
 $V[10][14] = 0.8825$; $V[11][14] = 0.9766$; $V[12][14] = 0.9654$; $V[13][14] = 0.8686$; $V[14][14]$
 $= 0.4814$; $V[15][14] = -0.2038$; $V[16][14] = -0.7782$; $V[17][14] = -0.9803$; $V[18][14]$
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 $= 0.6685$; $V[23][14] = 0.9685$; $V[24][14] = 1.0037$; $V[25][14] = 0.9819$; $V[0][15] =$
 0.9751 ; $V[1][15] = 0.7798$; $V[2][15] = 0.2234$; $V[3][15] = -0.5025$; $V[4][15] = -0.9268$;
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 $= 0.4151$; $V[10][15] = 0.8820$; $V[11][15] = 0.9882$; $V[12][15] = 0.9778$; $V[13][15] =$
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 $V[17][16] = -0.9885$; $V[18][16] = -1.0010$; $V[19][16] = -0.9653$; $V[20][16] = -0.6772$;

$V[21][16] = 0.0422$; $V[22][16] = 0.7339$; $V[23][16] = 0.9766$; $V[24][16] = 1.0003$; $V[25][16] = 0.9793$; $V[0][17] = 0.9196$; $V[1][17] = 0.5457$; $V[2][17] = -0.1520$; $V[3][17] = -0.7404$; $V[4][17] = -0.9644$; $V[5][17] = -1.0009$; $V[6][17] = -1.0009$; $V[7][17] = -0.8317$; $V[8][17] = -0.2139$; $V[9][17] = 0.5522$; $V[10][17] = 0.9232$; $V[11][17] = 0.9847$; $V[12][17] = 0.9604$; $V[13][17] = 0.8216$; $V[14][17] = 0.3378$; $V[15][17] = -0.3922$; $V[16][17] = -0.8583$; $V[17][17] = -0.9868$; $V[18][17] = -0.9975$; $V[19][17] = -0.9463$; $V[20][17] = -0.6150$; $V[21][17] = 0.1268$; $V[22][17] = 0.7786$; $V[23][17] = 0.9850$; $V[24][17] = 1.0015$; $V[25][17] = 0.9800$; $V[0][18] = 0.8173$; $V[1][18] = 0.3214$; $V[2][18] = -0.3936$; $V[3][18] = -0.8566$; $V[4][18] = -0.9865$; $V[5][18] = -1.0018$; $V[6][18] = -0.9856$; $V[7][18] = -0.7325$; $V[8][18] = -0.0736$; $V[9][18] = 0.6192$; $V[10][18] = 0.9266$; $V[11][18] = 0.9773$; $V[12][18] = 0.9523$; $V[13][18] = 0.8084$; $V[14][18] = 0.2763$; $V[15][18] = -0.4594$; $V[16][18] = -0.8959$; $V[17][18] = -0.9906$; $V[18][18] = -0.9988$; $V[19][18] = -0.9293$; $V[20][18] = -0.5428$; $V[21][18] = 0.2249$; $V[22][18] = 0.8276$; $V[23][18] = 0.9934$; $V[24][18] = 1.0022$; $V[25][18] = 0.9833$; $V[0][19] = 0.6442$; $V[1][19] = 0.0318$; $V[2][19] = -0.6315$; $V[3][19] = -0.9437$; $V[4][19] = -0.9994$; $V[5][19] = -1.0036$; $V[6][19] = -0.9547$; $V[7][19] = -0.6277$; $V[8][19] = 0.0365$; $V[9][19] = 0.6635$; $V[10][19] = 0.9280$; $V[11][19] = 0.9798$; $V[12][19] = 0.9533$; $V[13][19] = 0.7767$; $V[14][19] = 0.1949$; $V[15][19] = -0.5327$; $V[16][19] = -0.9225$; $V[17][19] = -0.9976$; $V[18][19] = -1.0013$; $V[19][19] = -0.9179$; $V[20][19] = -0.4610$; $V[21][19] = 0.3270$; $V[22][19] = 0.8631$; $V[23][19] = 0.9973$; $V[24][19] = 1.0025$; $V[25][19] = 0.9796$; $V[0][20] = 0.3505$; $V[1][20] = -0.2982$; $V[2][20] = -0.8151$; $V[3][20] = -0.9856$; $V[4][20] = -1.0022$; $V[5][20] = -1.0011$; $V[6][20] = -0.9054$; $V[7][20] = -0.5010$; $V[8][20] = 0.1743$; $V[9][20] = 0.7314$; $V[10][20] = 0.9433$; $V[11][20] = 0.9840$; $V[12][20] = 0.9515$; $V[13][20] = 0.7229$; $V[14][20] = 0.1042$; $V[15][20] = -0.5992$; $V[16][20] = -0.9428$; $V[17][20] = -1.0026$; $V[18][20] = -1.0026$; $V[19][20] = -0.9011$; $V[20][20] = -0.3712$; $V[21][20] = 0.4248$; $V[22][20] = 0.8960$; $V[23][20] = 0.9969$; $V[24][20] = 1.0029$; $V[25][20] = 0.9659$; $V[0][21] = -0.0838$; $V[1][21] = -0.6370$; $V[2][21] = -0.9280$; $V[3][21] = -1.0005$; $V[4][21] = -1.0015$; $V[5][21] = -0.9732$; $V[6][21] = -0.7993$; $V[7][21]$

= -0.3115; V[8][21] = 0.3614; V[9][21] = 0.8168; V[10][21] = 0.9592; V[11][21] = 0.9873;
 V[12][21] = 0.9438; V[13][21] = 0.6739; V[14][21] = 0.0195; V[15][21] = -0.6583; V[16][21]
 = -0.9573; V[17][21] = -0.9998; V[18][21] = -1.0034; V[19][21] = -0.8722; V[20][21]
 = -0.2783; V[21][21] = 0.5265; V[22][21] = 0.9298; V[23][21] = 0.9989; V[24][21] =
 1.0004; V[25][21] = 0.9508; V[0][22] = -0.5533; V[1][22] = -0.8802; V[2][22] = -0.9818;
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 = -0.0504; V[8][22] = 0.5403; V[9][22] = 0.8861; V[10][22] = 0.9793; V[11][22] = 0.9843;
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 = 0.2026; V[7][24] = 0.5844; V[8][24] = 0.8163; V[9][24] = 0.9275; V[10][24] = 0.9312;
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 1.0008 ; $V[13][44] = 0.9656$; $V[14][44] = 0.7763$; $V[15][44] = 0.2224$; $V[16][44] = -$
 0.5030 ; $V[17][44] = -0.9162$; $V[18][44] = -1.0020$; $V[19][44] = -1.0024$; $V[20][44] =$
 -0.9107 ; $V[21][44] = -0.4679$; $V[22][44] = 0.2692$; $V[23][44] = 0.8181$; $V[24][44] =$
 0.9903 ; $V[25][44] = 1.0014$; $V[0][45] = 0.9930$; $V[1][45] = 0.9716$; $V[2][45] = 0.7184$;
 $V[3][45] = 0.0239$; $V[4][45] = -0.6997$; $V[5][45] = -0.9793$; $V[6][45] = -1.0052$; $V[7][45]$
 $= -0.9908$; $V[8][45] = -0.7730$; $V[9][45] = -0.1079$; $V[10][45] = 0.6406$; $V[11][45] =$
 0.9670 ; $V[12][45] = 1.0022$; $V[13][45] = 0.9737$; $V[14][45] = 0.7957$; $V[15][45] = 0.2477$;
 $V[16][45] = -0.4962$; $V[17][45] = -0.9290$; $V[18][45] = -1.0023$; $V[19][45] = -1.0015$;
 $V[20][45] = -0.9055$; $V[21][45] = -0.4389$; $V[22][45] = 0.3124$; $V[23][45] = 0.8437$;
 $V[24][45] = 0.9966$; $V[25][45] = 1.0014$; $V[0][46] = 0.9951$; $V[1][46] = 0.9792$; $V[2][46]$
 $= 0.7594$; $V[3][46] = 0.0744$; $V[4][46] = -0.6593$; $V[5][46] = -0.9750$; $V[6][46] = -$
 1.0041 ; $V[7][46] = -0.9876$; $V[8][46] = -0.7855$; $V[9][46] = -0.1265$; $V[10][46] = 0.6304$;
 $V[11][46] = 0.9675$; $V[12][46] = 0.9988$; $V[13][46] = 0.9736$; $V[14][46] = 0.8212$; $V[15][46]$
 $= 0.2635$; $V[16][46] = -0.5098$; $V[17][46] = -0.9390$; $V[18][46] = -1.0018$; $V[19][46] =$
 -0.9997 ; $V[20][46] = -0.9022$; $V[21][46] = -0.4482$; $V[22][46] = 0.3248$; $V[23][46] =$
 0.8597 ; $V[24][46] = 0.9989$; $V[25][46] = 1.0004$; $V[0][47] = 0.9956$; $V[1][47] = 0.9842$;
 $V[2][47] = 0.7515$; $V[3][47] = 0.0637$; $V[4][47] = -0.6813$; $V[5][47] = -0.9789$; $V[6][47]$
 $= -1.0046$; $V[7][47] = -0.9805$; $V[8][47] = -0.7589$; $V[9][47] = -0.1180$; $V[10][47] =$
 0.6133 ; $V[11][47] = 0.9496$; $V[12][47] = 0.9933$; $V[13][47] = 0.9685$; $V[14][47] = 0.8268$;
 $V[15][47] = 0.2892$; $V[16][47] = -0.4874$; $V[17][47] = -0.9282$; $V[18][47] = -1.0040$;

$V[19][47] = -0.9964$; $V[20][47] = -0.9128$; $V[21][47] = -0.4645$; $V[22][47] = 0.3265$;
 $V[23][47] = 0.8692$; $V[24][47] = 0.9973$; $V[25][47] = 0.9994$; $V[0][48] = 0.9949$; $V[1][48]$
 $= 0.9867$; $V[2][48] = 0.7496$; $V[3][48] = 0.0515$; $V[4][48] = -0.6969$; $V[5][48] = -0.9848$;
 $V[6][48] = -1.0046$; $V[7][48] = -0.9800$; $V[8][48] = -0.7527$; $V[9][48] = -0.1239$; $V[10][48]$
 $= 0.5897$; $V[11][48] = 0.9286$; $V[12][48] = 0.9940$; $V[13][48] = 0.9819$; $V[14][48] =$
 0.8472 ; $V[15][48] = 0.3171$; $V[16][48] = -0.4715$; $V[17][48] = -0.9261$; $V[18][48] = -$
 1.0041 ; $V[19][48] = -1.0007$; $V[20][48] = -0.9163$; $V[21][48] = -0.4481$; $V[22][48] =$
 0.3582 ; $V[23][48] = 0.8896$; $V[24][48] = 0.9968$; $V[25][48] = 1.0000$; $V[0][49] = 0.9927$;
 $V[1][49] = 0.9835$; $V[2][49] = 0.7844$; $V[3][49] = 0.1163$; $V[4][49] = -0.6497$; $V[5][49]$
 $= -0.9786$; $V[6][49] = -1.0042$; $V[7][49] = -0.9876$; $V[8][49] = -0.7908$; $V[9][49] = -$
 0.1905 ; $V[10][49] = 0.5225$; $V[11][49] = 0.9077$; $V[12][49] = 0.9935$; $V[13][49] = 0.9891$;
 $V[14][49] = 0.8584$; $V[15][49] = 0.3031$; $V[16][49] = -0.4961$; $V[17][49] = -0.9375$;
 $V[18][49] = -1.0031$; $V[19][49] = -1.0036$; $V[20][49] = -0.9196$; $V[21][49] = -0.4386$;
 $V[22][49] = 0.3721$; $V[23][49] = 0.8954$; $V[24][49] = 0.9990$; $V[25][49] = 1.0006$; $V[0][50]$
 $= 0.9870$; $V[1][50] = 0.9834$; $V[2][50] = 0.8140$; $V[3][50] = 0.1893$; $V[4][50] = -0.5929$;
 $V[5][50] = -0.9705$; $V[6][50] = -1.0047$; $V[7][50] = -0.9915$; $V[8][50] = -0.8309$; $V[9][50]$
 $= -0.2543$; $V[10][50] = 0.4833$; $V[11][50] = 0.8952$; $V[12][50] = 0.9858$; $V[13][50] =$
 0.9813 ; $V[14][50] = 0.8389$; $V[15][50] = 0.2841$; $V[16][50] = -0.5200$; $V[17][50] = -$
 0.9362 ; $V[18][50] = -1.0021$; $V[19][50] = -1.0025$; $V[20][50] = -0.9131$; $V[21][50] = -$
 0.4337 ; $V[22][50] = 0.3725$; $V[23][50] = 0.8895$; $V[24][50] = 1.0012$; $V[25][50] = 1.0004$;
 $V[0][51] = 0.9849$; $V[1][51] = 0.9828$; $V[2][51] = 0.8140$; $V[3][51] = 0.2276$; $V[4][51]$
 $= -0.5667$; $V[5][51] = -0.9630$; $V[6][51] = -1.0049$; $V[7][51] = -0.9912$; $V[8][51] = -$
 0.8322 ; $V[9][51] = -0.2476$; $V[10][51] = 0.4969$; $V[11][51] = 0.9018$; $V[12][51] = 0.9835$;
 $V[13][51] = 0.9840$; $V[14][51] = 0.8409$; $V[15][51] = 0.2827$; $V[16][51] = -0.5014$; $V[17][51]$
 $= -0.9233$; $V[18][51] = -1.0024$; $V[19][51] = -1.0005$; $V[20][51] = -0.8955$; $V[21][51] = -$
 0.4052 ; $V[22][51] = 0.3990$; $V[23][51] = 0.8958$; $V[24][51] = 1.0008$; $V[25][51] = 1.0005$;

B.0.2 Defect Order = 2

$if(m_i n d e x == 1 \&\& (m_{p o s . x} - 0.25 * p i t c h) > (-2.25 * p i t c h) \&\& (m_{p o s . x} - 0.25 * p i t c h) < (2.25 * p i t c h) \&\& m_{p o s . y} > (-2.0 * p i t c h) \&\& m_{p o s . y} < (2.0 * p i t c h))$

FX[0][0]=9.4627; FX[1][0]=4.5195; FX[2][0]=0.5615; FX[3][0]=0.0780; FX[4][0] = -1.0476; FX[5][0] = -6.0438; FX[6][0] = -10.5300; FX[7][0] = -6.3533; FX[8][0] = -1.0331; FX[9][0] = -0.0498; FX[10][0] = 0.4365; FX[11][0] = 4.1063; FX[12][0] = 9.6133; FX[13][0] = 8.0426; FX[14][0] = 2.4972; FX[15][0] = 0.2093; FX[16][0] = -0.3318; FX[17][0] = -2.1742; FX[18][0] = -7.4123; FX[19][0] = -9.5630; FX[20][0] = -4.7433; FX[21][0] = -0.6814; FX[22][0] = 0.0272; FX[23][0] = 1.3852; FX[24][0] = 6.5103; FX[25][0] = 10.0727; FX[26][0] = 5.9131; FX[27][0] = 1.0816; FX[28][0] = -0.0008; FX[29][0] = -0.5516; FX[0][1] = 9.4672; FX[1][1] = 4.4473; FX[2][1] = 0.5645; FX[3][1] = 0.0668; FX[4][1] = -1.0561; FX[5][1] = -6.0078; FX[6][1] = -10.5829; FX[7][1] = -6.3848; FX[8][1] = -0.9824; FX[9][1] = -0.0342; FX[10][1] = 0.2837; FX[11][1] = 3.8426; FX[12][1] = 9.6592; FX[13][1] = 8.3689; FX[14][1] = 2.5798; FX[15][1] = 0.2845; FX[16][1] = -0.3331; FX[17][1] = -2.2470; FX[18][1] = -7.9399; FX[19][1] = -9.6178; FX[20][1] = -4.3837; FX[21][1] = -0.4838; FX[22][1] = 0.0000; FX[23][1] = 1.0935; FX[24][1] = 6.4750; FX[25][1] = 10.0299; FX[26][1] = 6.0631; FX[27][1] = 1.2760; FX[28][1] = 0.0940; FX[29][1] = -0.4044; FX[0][2] = 9.9473; FX[1][2] = 4.9646; FX[2][2] = 0.8672; FX[3][2] = 0.0394; FX[4][2] = -0.9184; FX[5][2] = -5.8268; FX[6][2] = -10.3816; FX[7][2] = -6.7035; FX[8][2] = -1.1588; FX[9][2] = -0.0068; FX[10][2] = 0.3320; FX[11][2] = 3.7961; FX[12][2] = 9.6369; FX[13][2] = 8.6192; FX[14][2] = 2.5183; FX[15][2] = 0.1813; FX[16][2] = -0.3298; FX[17][2] = -2.2246; FX[18][2] = -8.2869; FX[19][2] = -9.8912; FX[20][2] = -4.0061; FX[21][2] = -0.3099; FX[22][2] = -0.0329; FX[23][2] = 1.0211; FX[24][2] = 6.4833; FX[25][2] = 10.0018; FX[26][2] = 6.0355; FX[27][2] = 1.3745; FX[28][2] = 0.1185; FX[29][2] = -0.3220; FX[0][3] = 10.0823; FX[1][3] = 5.3164; FX[2][3] = 1.1039; FX[3][3] = 0.0742; FX[4][3] = -0.8375; FX[5][3] = -5.3953; FX[6][3] = -10.1287; FX[7][3] = -7.0220; FX[8][3] = -1.4993; FX[9][3] = -0.1077; FX[10][3] = 0.4200;

FX[11][3] = 4.1752; FX[12][3] = 9.9931; FX[13][3] = 8.2252; FX[14][3] = 2.2173; FX[15][3] = 0.0644; FX[16][3] = -0.2273; FX[17][3] = -2.2925; FX[18][3] = -8.2625; FX[19][3] = -9.9836; FX[20][3] = -3.8597; FX[21][3] = -0.3849; FX[22][3] = -0.0475; FX[23][3] = 1.1211; FX[24][3] = 6.4804; FX[25][3] = 10.1187; FX[26][3] = 5.8650; FX[27][3] = 1.4035; FX[28][3] = 0.0392; FX[29][3] = -0.3570; FX[0][4] = 10.3345; FX[1][4] = 5.2102; FX[2][4] = 1.1997; FX[3][4] = 0.1196; FX[4][4] = -0.9783; FX[5][4] = -5.3696; FX[6][4] = -9.8221; FX[7][4] = -7.1471; FX[8][4] = -1.5584; FX[9][4] = -0.0984; FX[10][4] = 0.4311; FX[11][4] = 4.5611; FX[12][4] = 10.0562; FX[13][4] = 7.9162; FX[14][4] = 1.9924; FX[15][4] = 0.0942; FX[16][4] = -0.1250; FX[17][4] = -2.2656; FX[18][4] = -8.1858; FX[19][4] = -9.7884; FX[20][4] = -4.1011; FX[21][4] = -0.5231; FX[22][4] = -0.0470; FX[23][4] = 1.0596; FX[24][4] = 6.4998; FX[25][4] = 10.3627; FX[26][4] = 6.0294; FX[27][4] = 1.1061; FX[28][4] = -0.0134; FX[29][4] = -0.3296; FX[0][5] = 9.9386; FX[1][5] = 5.3627; FX[2][5] = 1.2637; FX[3][5] = 0.1178; FX[4][5] = -0.9525; FX[5][5] = -5.3778; FX[6][5] = -10.1447; FX[7][5] = -7.0344; FX[8][5] = -1.4258; FX[9][5] = 0.0049; FX[10][5] = 0.2621; FX[11][5] = 4.3814; FX[12][5] = 9.8818; FX[13][5] = 7.9408; FX[14][5] = 2.3416; FX[15][5] = 0.2063; FX[16][5] = -0.0729; FX[17][5] = -2.2155; FX[18][5] = -8.2685; FX[19][5] = -9.9424; FX[20][5] = -3.9553; FX[21][5] = -0.5517; FX[22][5] = -0.0056; FX[23][5] = 1.0257; FX[24][5] = 6.5915; FX[25][5] = 10.6755; FX[26][5] = 5.9794; FX[27][5] = 0.7925; FX[28][5] = -0.0382; FX[29][5] = -0.3772; FX[0][6] = 9.6415; FX[1][6] = 5.4157; FX[2][6] = 1.1729; FX[3][6] = 0.1825; FX[4][6] = -0.8075; FX[5][6] = -5.2928; FX[6][6] = -10.5003; FX[7][6] = -6.9685; FX[8][6] = -1.3753; FX[9][6] = -0.0450; FX[10][6] = 0.2245; FX[11][6] = 4.5182; FX[12][6] = 9.7537; FX[13][6] = 7.8906; FX[14][6] = 2.3411; FX[15][6] = 0.1870; FX[16][6] = -0.2201; FX[17][6] = -2.3739; FX[18][6] = -8.4363; FX[19][6] = -9.5657; FX[20][6] = -3.8436; FX[21][6] = -0.4552; FX[22][6] = -0.0237; FX[23][6] = 1.2478; FX[24][6] = 7.1321; FX[25][6] = 10.5229; FX[26][6] = 5.5916; FX[27][6] = 0.5717; FX[28][6] = -0.0199; FX[29][6] = -0.4099; FX[0][7] = 9.5827; FX[1][7] = 5.3509; FX[2][7] =

1.1775; FX[3][7] = 0.2430; FX[4][7] = -0.5329; FX[5][7] = -5.1858; FX[6][7] = -10.7860;
 FX[7][7] = -7.0715; FX[8][7] = -1.3665; FX[9][7] = -0.1496; FX[10][7] = 0.5153; FX[11][7] =
 = 5.1124; FX[12][7] = 9.9404; FX[13][7] = 7.2830; FX[14][7] = 1.9227; FX[15][7] =
 -0.0016; FX[16][7] = -0.5052; FX[17][7] = -2.8778; FX[18][7] = -8.6056; FX[19][7] =
 -8.9314; FX[20][7] = -3.4211; FX[21][7] = -0.4165; FX[22][7] = -0.0034; FX[23][7] =
 1.5032; FX[24][7] = 7.5337; FX[25][7] = 10.4856; FX[26][7] = 5.0463; FX[27][7] =
 0.4685; FX[28][7] = -0.0479; FX[29][7] = -0.5251; FX[0][8] = 9.7190; FX[1][8] = 5.4157;
 FX[2][8] = 1.2677; FX[3][8] = 0.1661; FX[4][8] = -0.5541; FX[5][8] = -5.4128; FX[6][8]
 = -10.7670; FX[7][8] = -6.9826; FX[8][8] = -1.2966; FX[9][8] = -0.1213; FX[10][8] =
 0.7640; FX[11][8] = 5.3531; FX[12][8] = 10.0641; FX[13][8] = 7.0492; FX[14][8] =
 1.7369; FX[15][8] = -0.1004; FX[16][8] = -0.5824; FX[17][8] = -3.5785; FX[18][8] =
 -9.0084; FX[19][8] = -8.5188; FX[20][8] = -2.8891; FX[21][8] = -0.2577; FX[22][8]
 = 0.0467; FX[23][8] = 1.5927; FX[24][8] = 8.0289; FX[25][8] = 10.3175; FX[26][8]
 = 4.6889; FX[27][8] = 0.4012; FX[28][8] = -0.0961; FX[29][8] = -0.6390; FX[0][9] =
 9.4118; FX[1][9] = 5.2819; FX[2][9] = 1.1521; FX[3][9] = 0.1118; FX[4][9] = -0.8746;
 FX[5][9] = -5.8782; FX[6][9] = -10.4941; FX[7][9] = -6.5463; FX[8][9] = -1.2354; FX[9][9]
 = -0.0767; FX[10][9] = 0.7826; FX[11][9] = 5.3785; FX[12][9] = 9.8815; FX[13][9] =
 7.1503; FX[14][9] = 1.7854; FX[15][9] = 0.0371; FX[16][9] = -0.6796; FX[17][9] = -
 3.7327; FX[18][9] = -9.2997; FX[19][9] = -8.4319; FX[20][9] = -2.7082; FX[21][9] =
 -0.1217; FX[22][9] = 0.0272; FX[23][9] = 1.8979; FX[24][9] = 8.3873; FX[25][9] =
 10.2937; FX[26][9] = 4.1761; FX[27][9] = 0.2945; FX[28][9] = -0.0740; FX[29][9] =
 -0.3030; FX[0][10] = 9.2120; FX[1][10] = 4.8932; FX[2][10] = 0.9444; FX[3][10] =
 0.0537; FX[4][10] = -0.9700; FX[5][10] = -6.3148; FX[6][10] = -10.2273; FX[7][10] =
 -6.0981; FX[8][10] = -1.3101; FX[9][10] = -0.1434; FX[10][10] = 0.7672; FX[11][10] =
 5.5001; FX[12][10] = 9.7922; FX[13][10] = 6.9217; FX[14][10] = 1.8984; FX[15][10] =
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 -8.5740; FX[20][10] = -2.7209; FX[21][10] = -0.0826; FX[22][10] = -0.0171; FX[23][10]

= 2.2070; FX[24][10] = 8.8170; FX[25][10] = 10.0292; FX[26][10] = 3.7188; FX[27][10] = 0.3135; FX[28][10] = -0.0770; FX[29][10] = -0.2692; FX[0][11] = 9.4416; FX[1][11] = 4.5277; FX[2][11] = 0.9084; FX[3][11] = 0.0578; FX[4][11] = -0.8507; FX[5][11] = -6.1433; FX[6][11] = -10.3188; FX[7][11] = -6.3200; FX[8][11] = -1.2980; FX[9][11] = -0.1493; FX[10][11] = 0.6863; FX[11][11] = 5.5249; FX[12][11] = 9.9593; FX[13][11] = 6.8211; FX[14][11] = 1.7702; FX[15][11] = 0.1679; FX[16][11] = -0.7426; FX[17][11] = -3.7375; FX[18][11] = -8.8923; FX[19][11] = -8.5676; FX[20][11] = -2.8302; FX[21][11] = -0.1315; FX[22][11] = 0.0013; FX[23][11] = 2.4085; FX[24][11] = 8.9148; FX[25][11] = 9.8761; FX[26][11] = 3.5386; FX[27][11] = 0.3157; FX[28][11] = -0.0845; FX[29][11] = -0.4891; FX[0][12] = 9.3112; FX[1][12] = 4.2599; FX[2][12] = 0.8401; FX[3][12] = 0.0466; FX[4][12] = -0.6385; FX[5][12] = -5.8019; FX[6][12] = -10.5256; FX[7][12] = -6.7549; FX[8][12] = -1.2684; FX[9][12] = -0.0740; FX[10][12] = 0.7619; FX[11][12] = 5.5772; FX[12][12] = 10.1284; FX[13][12] = 6.6331; FX[14][12] = 1.5379; FX[15][12] = 0.1895; FX[16][12] = -0.7024; FX[17][12] = -4.1471; FX[18][12] = -9.0664; FX[19][12] = -7.8998; FX[20][12] = -2.7906; FX[21][12] = -0.1824; FX[22][12] = 0.1201; FX[23][12] = 2.6113; FX[24][12] = 8.8068; FX[25][12] = 9.6590; FX[26][12] = 3.4856; FX[27][12] = 0.3600; FX[28][12] = -0.1413; FX[29][12] = -1.1508; FX[0][13] = 8.6779; FX[1][13] = 3.5900; FX[2][13] = 0.6404; FX[3][13] = 0.0555; FX[4][13] = -0.6981; FX[5][13] = -5.6939; FX[6][13] = -10.6396; FX[7][13] = -6.7327; FX[8][13] = -1.2550; FX[9][13] = -0.0370; FX[10][13] = 1.0445; FX[11][13] = 5.7947; FX[12][13] = 10.2398; FX[13][13] = 6.4017; FX[14][13] = 1.1017; FX[15][13] = 0.1270; FX[16][13] = -0.8322; FX[17][13] = -4.6739; FX[18][13] = -8.9941; FX[19][13] = -7.4751; FX[20][13] = -2.4941; FX[21][13] = -0.2243; FX[22][13] = 0.2969; FX[23][13] = 2.8479; FX[24][13] = 8.7031; FX[25][13] = 9.2963; FX[26][13] = 3.5381; FX[27][13] = 0.4018; FX[28][13] = -0.2130; FX[29][13] = -1.4858; FX[0][14] = 7.8645; FX[1][14] = 2.9363; FX[2][14] = 0.3326; FX[3][14] = 0.0134; FX[4][14] = -1.2824; FX[5][14] = -6.1762; FX[6][14] = -10.1345; FX[7][14] = -6.2592; FX[8][14] = -1.1458; FX[9][14] = -0.0403; FX[10][14] = 1.3201; FX[11][14] =

6.3503; FX[12][14] = 10.1282; FX[13][14] = 5.9622; FX[14][14] = 0.9924; FX[15][14] =
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 = 10.0547; FX[26][31] = 5.6436; FX[27][31] = 0.9475; FX[28][31] = 0.0785; FX[29][31]
 = -0.0941; FX[0][32] = 2.6259; FX[1][32] = 7.2662; FX[2][32] = 8.8797; FX[3][32] =
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-0.2495; FX[12][32] = 1.4616; FX[13][32] = 5.2062; FX[14][32] = 7.8324; FX[15][32] =
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 -7.5406; FX[20][32] = -6.6120; FX[21][32] = -2.5536; FX[22][32] = -0.2212; FX[23][32]
 = 1.0336; FX[24][32] = 5.7639; FX[25][32] = 10.0048; FX[26][32] = 6.6767; FX[27][32]
 = 1.3437; FX[28][32] = 0.1098; FX[29][32] = -0.1501; FX[0][33] = 4.6366; FX[1][33]
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 = 9.5348; FX[26][33] = 7.4744; FX[27][33] = 1.9164; FX[28][33] = 0.0499; FX[29][33]
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 = 6.7537; FX[2][45] = 1.5107; FX[3][45] = 0.0807; FX[4][45] = -0.2170; FX[5][45] =
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= 1.0995; FX[24][46] = 5.7271; FX[25][46] = 9.8899; FX[26][46] = 6.7017; FX[27][46]
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 -0.2832; FY[18][41] = -0.8097; FY[19][41] = -0.8894; FY[20][41] = -0.2732; FY[21][41]
 = 0.0615; FY[22][41] = 0.0021; FY[23][41] = -0.1474; FY[24][41] = 0.0450; FY[25][41]
 = 0.4383; FY[26][41] = 0.4291; FY[27][41] = 0.1396; FY[28][41] = -0.0016; FY[29][41]

= -0.0036; FY[0][42] = 0.1155; FY[1][42] = 0.0217; FY[2][42] = -0.0997; FY[3][42] =
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 = 0.0290; FY[20][46] = -0.1267; FY[21][46] = -0.0308; FY[22][46] = 0.0101; FY[23][46]
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 = 0.1795; FY[20][48] = -0.0305; FY[21][48] = -0.0162; FY[22][48] = -0.0179; FY[23][48]
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 = 0.1194; FY[2][49] = -0.0966; FY[3][49] = -0.1338; FY[4][49] = -0.0357; FY[5][49]

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= -0.3651; V[19][31] = 0.1223; V[20][31] = 0.6505; V[21][31] = 0.9337; V[22][31] = 0.9965; V[23][31] = 0.9637; V[24][31] = 0.6637; V[25][31] = -0.0718; V[26][31] = -0.7392; V[27][31] = -0.9716; V[28][31] = -0.9954; V[29][31] = -0.9979; V[0][32] = 0.9209; V[1][32] = 0.5339; V[2][32] = -0.1629; V[3][32] = -0.7485; V[4][32] = -0.9800; V[5][32] = -1.0015; V[6][32] = -0.9838; V[7][32] = -0.8328; V[8][32] = -0.2991; V[9][32] = 0.4317; V[10][32] = 0.8609; V[11][32] = 0.9728; V[12][32] = 0.9327; V[13][32] = 0.6809; V[14][32] = 0.1254; V[15][32] = -0.4443; V[16][32] = -0.7311; V[17][32] = -0.7607; V[18][32] = -0.5563; V[19][32] = -0.0709; V[20][32] = 0.5334; V[21][32] = 0.9058; V[22][32] = 0.9947; V[23][32] = 0.9725; V[24][32] = 0.7303; V[25][32] = 0.0577; V[26][32] = -0.6636; V[27][32] = -0.9596; V[28][32] = -0.9955; V[29][32] = -0.9958; V[0][33] = 0.7988; V[1][33] = 0.2517; V[2][33] = -0.4455; V[3][33] = -0.8721; V[4][33] = -0.9906; V[5][33] = -0.9990; V[6][33] = -0.9557; V[7][33] = -0.6581; V[8][33] = 0.0458; V[9][33] = 0.7111; V[10][33] = 0.9655; V[11][33] = 1.0055; V[12][33] = 0.9672; V[13][33] = 0.6896; V[14][33] = 0.0579; V[15][33] = -0.5749; V[16][33] = -0.8554; V[17][33] = -0.8726; V[18][33] = -0.6972; V[19][33] = -0.2131; V[20][33] = 0.4469; V[21][33] = 0.8850; V[22][33] = 0.9924; V[23][33] = 0.9654; V[24][33] = 0.7574; V[25][33] = 0.1521; V[26][33] = -0.5833; V[27][33] = -0.9516; V[28][33] = -0.9988; V[29][33] = -0.9972; V[0][34] = 0.6484; V[1][34] = -0.0079; V[2][34] = -0.6474; V[3][34] = -0.9357; V[4][34] = -0.9971; V[5][34] = -1.0000; V[6][34] = -0.9126; V[7][34] = -0.4394; V[8][34] = 0.3532; V[9][34] = 0.8672; V[10][34] = 0.9964; V[11][34] = 1.0051; V[12][34] = 0.9590; V[13][34] = 0.6152; V[14][34] = -0.1197; V[15][34] = -0.7408; V[16][34] = -0.9411; V[17][34] = -0.9375; V[18][34] = -0.7928; V[19][34] = -0.3137; V[20][34] = 0.3981; V[21][34] = 0.8766; V[22][34] = 0.9951; V[23][34] = 0.9747; V[24][34] = 0.7964; V[25][34] = 0.2250; V[26][34] = -0.5305; V[27][34] = -0.9308; V[28][34] = -0.9967; V[29][34] = -0.9965; V[0][35] = 0.5155; V[1][35] = -0.2031; V[2][35] = -0.7742; V[3][35] = -0.9729; V[4][35] = -0.9997; V[5][35] = -0.9979; V[6][35] = -0.8336; V[7][35] = -0.1901; V[8][35] = 0.5975; V[9][35] = 0.9573; V[10][35] = 1.0037; V[11][35] = 1.0024;

$V[12][35] = 0.9381$; $V[13][35] = 0.5140$; $V[14][35] = -0.2859$; $V[15][35] = -0.8564$;
 $V[16][35] = -0.9870$; $V[17][35] = -0.9676$; $V[18][35] = -0.8209$; $V[19][35] = -0.3258$;
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 $= 0.8360$; $V[25][35] = 0.2796$; $V[26][35] = -0.4959$; $V[27][35] = -0.9221$; $V[28][35]$
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$V[5][39] = -0.9051$; $V[6][39] = -0.4297$; $V[7][39] = 0.3938$; $V[8][39] = 0.9091$; $V[9][39] = 1.0015$; $V[10][39] = 1.0024$; $V[11][39] = 0.9597$; $V[12][39] = 0.6238$; $V[13][39] = -0.1380$; $V[14][39] = -0.7939$; $V[15][39] = -0.9865$; $V[16][39] = -0.9952$; $V[17][39] = -0.9744$; $V[18][39] = -0.7647$; $V[19][39] = -0.1139$; $V[20][39] = 0.6302$; $V[21][39] = 0.9592$; $V[22][39] = 1.0026$; $V[23][39] = 0.9895$; $V[24][39] = 0.8266$; $V[25][39] = 0.2366$; $V[26][39] = -0.5480$; $V[27][39] = -0.9456$; $V[28][39] = -1.0009$; $V[29][39] = -0.9951$; $V[0][40] = 0.0312$; $V[1][40] = -0.6397$; $V[2][40] = -0.9409$; $V[3][40] = -0.9983$; $V[4][40] = -0.9909$; $V[5][40] = -0.8724$; $V[6][40] = -0.3676$; $V[7][40] = 0.4264$; $V[8][40] = 0.9138$; $V[9][40] = 1.0031$; $V[10][40] = 1.0033$; $V[11][40] = 0.9389$; $V[12][40] = 0.5363$; $V[13][40] = -0.2425$; $V[14][40] = -0.8290$; $V[15][40] = -0.9866$; $V[16][40] = -0.9920$; $V[17][40] = -0.9711$; $V[18][40] = -0.7172$; $V[19][40] = -0.0200$; $V[20][40] = 0.6858$; $V[21][40] = 0.9708$; $V[22][40] = 1.0029$; $V[23][40] = 0.9823$; $V[24][40] = 0.8048$; $V[25][40] = 0.2138$; $V[26][40] = -0.5511$; $V[27][40] = -0.9427$; $V[28][40] = -0.9997$; $V[29][40] = -0.9963$; $V[0][41] = -0.0247$; $V[1][41] = -0.6902$; $V[2][41] = -0.9586$; $V[3][41] = -0.9965$; $V[4][41] = -0.9820$; $V[5][41] = -0.8298$; $V[6][41] = -0.3041$; $V[7][41] = 0.4581$; $V[8][41] = 0.9119$; $V[9][41] = 1.0041$; $V[10][41] = 1.0035$; $V[11][41] = 0.9389$; $V[12][41] = 0.4991$; $V[13][41] = -0.3112$; $V[14][41] = -0.8700$; $V[15][41] = -0.9976$; $V[16][41] = -0.9969$; $V[17][41] = -0.9583$; $V[18][41] = -0.6559$; $V[19][41] = 0.0552$; $V[20][41] = 0.7120$; $V[21][41] = 0.9720$; $V[22][41] = 1.0017$; $V[23][41] = 0.9858$; $V[24][41] = 0.7821$; $V[25][41] = 0.1670$; $V[26][41] = -0.5858$; $V[27][41] = -0.9473$; $V[28][41] = -0.9988$; $V[29][41] = -0.9964$; $V[0][42] = -0.0603$; $V[1][42] = -0.7213$; $V[2][42] = -0.9616$; $V[3][42] = -0.9905$; $V[4][42] = -0.9682$; $V[5][42] = -0.7914$; $V[6][42] = -0.2439$; $V[7][42] = 0.4918$; $V[8][42] = 0.9124$; $V[9][42] = 1.0029$; $V[10][42] = 1.0044$; $V[11][42] = 0.9357$; $V[12][42] = 0.4850$; $V[13][42] = -0.3462$; $V[14][42] = -0.8954$; $V[15][42] = -1.0047$; $V[16][42] = -0.9993$; $V[17][42] = -0.9371$; $V[18][42] = -0.5943$; $V[19][42] = 0.1248$; $V[20][42] = 0.7351$; $V[21][42] = 0.9653$; $V[22][42] = 1.0023$; $V[23][42] = 0.9944$; $V[24][42] = 0.7919$; $V[25][42] = 0.1446$; $V[26][42] = -0.6041$; $V[27][42] = -0.9565$; $V[28][42] =$

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 $= 1.0007$; $V[11][47] = 0.9179$; $V[12][47] = 0.4957$; $V[13][47] = -0.2783$; $V[14][47] =$
 -0.8455 ; $V[15][47] = -0.9918$; $V[16][47] = -0.9841$; $V[17][47] = -0.9310$; $V[18][47] = -$
 0.6140 ; $V[19][47] = 0.1168$; $V[20][47] = 0.7751$; $V[21][47] = 0.9924$; $V[22][47] = 1.0017$;
 $V[23][47] = 0.9772$; $V[24][47] = 0.7483$; $V[25][47] = 0.0760$; $V[26][47] = -0.6541$; $V[27][47]$
 $= -0.9667$; $V[28][47] = -1.0008$; $V[29][47] = -0.9986$; $V[0][48] = 0.2728$; $V[1][48] = -$
 0.4599 ; $V[2][48] = -0.8766$; $V[3][48] = -0.9865$; $V[4][48] = -1.0005$; $V[5][48] = -0.9203$;
 $V[6][48] = -0.4446$; $V[7][48] = 0.3644$; $V[8][48] = 0.9002$; $V[9][48] = 1.0044$; $V[10][48]$
 $= 1.0005$; $V[11][48] = 0.9090$; $V[12][48] = 0.4785$; $V[13][48] = -0.2701$; $V[14][48] =$
 -0.8176 ; $V[15][48] = -0.9804$; $V[16][48] = -0.9913$; $V[17][48] = -0.9381$; $V[18][48] = -$
 0.6363 ; $V[19][48] = 0.0899$; $V[20][48] = 0.7569$; $V[21][48] = 0.9913$; $V[22][48] = 1.0031$;
 $V[23][48] = 0.9816$; $V[24][48] = 0.7504$; $V[25][48] = 0.0631$; $V[26][48] = -0.6802$; $V[27][48]$
 $= -0.9733$; $V[28][48] = -0.9992$; $V[29][48] = -0.9995$; $V[0][49] = 0.2507$; $V[1][49] = -$
 0.4735 ; $V[2][49] = -0.8676$; $V[3][49] = -0.9793$; $V[4][49] = -0.9988$; $V[5][49] = -0.9276$;
 $V[6][49] = -0.4882$; $V[7][49] = 0.3192$; $V[8][49] = 0.8861$; $V[9][49] = 1.0034$; $V[10][49]$
 $= 1.0015$; $V[11][49] = 0.9246$; $V[12][49] = 0.5210$; $V[13][49] = -0.2071$; $V[14][49] =$
 -0.7773 ; $V[15][49] = -0.9744$; $V[16][49] = -0.9966$; $V[17][49] = -0.9568$; $V[18][49] = -$
 0.6497 ; $V[19][49] = 0.1019$; $V[20][49] = 0.7749$; $V[21][49] = 0.9933$; $V[22][49] = 1.0038$;
 $V[23][49] = 0.9889$; $V[24][49] = 0.7409$; $V[25][49] = 0.0378$; $V[26][49] = -0.7029$; $V[27][49]$
 $= -0.9767$; $V[28][49] = -1.0011$; $V[29][49] = -0.9991$; $V[0][50] = 0.2723$; $V[1][50] = -$
 0.4431 ; $V[2][50] = -0.8417$; $V[3][50] = -0.9648$; $V[4][50] = -0.9963$; $V[5][50] = -0.9360$;
 $V[6][50] = -0.5484$; $V[7][50] = 0.2417$; $V[8][50] = 0.8526$; $V[9][50] = 1.0036$; $V[10][50]$
 $= 1.0013$; $V[11][50] = 0.9486$; $V[12][50] = 0.5834$; $V[13][50] = -0.1405$; $V[14][50] =$
 -0.7464 ; $V[15][50] = -0.9640$; $V[16][50] = -0.9928$; $V[17][50] = -0.9451$; $V[18][50] = -$

0.6308; V[19][50] = 0.1270; V[20][50] = 0.7936; V[21][50] = 0.9906; V[22][50] = 1.0035;
V[23][50] = 0.9865; V[24][50] = 0.7430; V[25][50] = 0.0395; V[26][50] = -0.6935; V[27][50]
= -0.9774; V[28][50] = -1.0020; V[29][50] = -0.9982; V[0][51] = 0.3464; V[1][51] = -
0.3590; V[2][51] = -0.8067; V[3][51] = -0.9612; V[4][51] = -0.9956; V[5][51] = -0.9374;
V[6][51] = -0.5829; V[7][51] = 0.1891; V[8][51] = 0.8356; V[9][51] = 1.0033; V[10][51]
= 1.0008; V[11][51] = 0.9509; V[12][51] = 0.6024; V[13][51] = -0.1335; V[14][51] =
-0.7524; V[15][51] = -0.9612; V[16][51] = -0.9894; V[17][51] = -0.9443; V[18][51] = -
0.6178; V[19][51] = 0.1325; V[20][51] = 0.7785; V[21][51] = 0.9863; V[22][51] = 1.0040;
V[23][51] = 0.9801; V[24][51] = 0.7156; V[25][51] = 0.0139; V[26][51] = -0.7036; V[27][51]
= -0.9783; V[28][51] = -1.0020; V[29][51] = -0.9982;

B.0.3 Defect Order = 3

$if(m_{index} == 1 \&\& m_{pos.x} > (-2.5 * pitch) \&\& m_{pos.x} < (2.5 * pitch) \&\& m_{pos.y} >$
 $(-2.0 * pitch) \&\& m_{pos.y} < (2.0 * pitch))$

FX[0][0] = -0.0033; FX[1][0] = 1.3919; FX[2][0] = 6.1294; FX[3][0] = 9.9672; FX[4][0]
= 6.1569; FX[5][0] = 1.2316; FX[6][0] = 0.1263; FX[7][0] = -0.5307; FX[8][0] = -4.4950;
FX[9][0] = -10.0309; FX[10][0] = -8.0283; FX[11][0] = -1.9051; FX[12][0] = -0.0693;
FX[13][0] = 0.2226; FX[14][0] = 2.8057; FX[15][0] = 8.6573; FX[16][0] = 9.1751; FX[17][0]
= 3.5698; FX[18][0] = 0.4761; FX[19][0] = -0.2148; FX[20][0] = -1.3422; FX[21][0]
= -5.9147; FX[22][0] = -9.7698; FX[23][0] = -6.3524; FX[24][0] = -1.3269; FX[25][0]
= -0.0124; FX[26][0] = 0.8635; FX[27][0] = 5.0336; FX[28][0] = 9.8871; FX[29][0]
= 7.3519; FX[30][0] = 1.8253; FX[31][0] = 0.0598; FX[32][0] = -0.4623; FX[0][1] =
0.0398; FX[1][1] = 1.5403; FX[2][1] = 6.3857; FX[3][1] = 9.9113; FX[4][1] = 5.9894;
FX[5][1] = 0.9801; FX[6][1] = 0.1672; FX[7][1] = -0.5726; FX[8][1] = -4.4250; FX[9][1]
= -10.1169; FX[10][1] = -8.0094; FX[11][1] = -1.8572; FX[12][1] = -0.1065; FX[13][1]
= 0.1220; FX[14][1] = 2.6043; FX[15][1] = 8.6188; FX[16][1] = 9.3177; FX[17][1] =
3.7730; FX[18][1] = 0.4937; FX[19][1] = -0.1227; FX[20][1] = -1.5193; FX[21][1] =

-6.1357; FX[22][1] = -9.8808; FX[23][1] = -6.0599; FX[24][1] = -1.1748; FX[25][1] = -0.0351; FX[26][1] = 0.6227; FX[27][1] = 4.8415; FX[28][1] = 9.8903; FX[29][1] = 7.5407; FX[30][1] = 1.9711; FX[31][1] = 0.1917; FX[32][1] = -0.3589; FX[0][2] = 0.0428; FX[1][2] = 1.1862; FX[2][2] = 6.1138; FX[3][2] = 10.0526; FX[4][2] = 6.1714; FX[5][2] = 1.2861; FX[6][2] = 0.1076; FX[7][2] = -0.4849; FX[8][2] = -4.3315; FX[9][2] = -10.0121; FX[10][2] = -8.2363; FX[11][2] = -1.9264; FX[12][2] = -0.0375; FX[13][2] = 0.0925; FX[14][2] = 2.3832; FX[15][2] = 8.5043; FX[16][2] = 9.6529; FX[17][2] = 3.9202; FX[18][2] = 0.4913; FX[19][2] = -0.1680; FX[20][2] = -1.4063; FX[21][2] = -6.5481; FX[22][2] = -10.3084; FX[23][2] = -5.7499; FX[24][2] = -0.8511; FX[25][2] = -0.0213; FX[26][2] = 0.4513; FX[27][2] = 4.8320; FX[28][2] = 9.8667; FX[29][2] = 7.5235; FX[30][2] = 2.1088; FX[31][2] = 0.2859; FX[32][2] = -0.2429; FX[0][3] = -0.0456; FX[1][3] = 0.6865; FX[2][3] = 5.5394; FX[3][3] = 10.1673; FX[4][3] = 6.7525; FX[5][3] = 1.6747; FX[6][3] = 0.1341; FX[7][3] = -0.4107; FX[8][3] = -4.0302; FX[9][3] = -9.7108; FX[10][3] = -8.3986; FX[11][3] = -2.3158; FX[12][3] = -0.1205; FX[13][3] = 0.1326; FX[14][3] = 2.5494; FX[15][3] = 8.7721; FX[16][3] = 9.5915; FX[17][3] = 3.7217; FX[18][3] = 0.3376; FX[19][3] = -0.1849; FX[20][3] = -1.3724; FX[21][3] = -6.7188; FX[22][3] = -10.4496; FX[23][3] = -5.6144; FX[24][3] = -0.7065; FX[25][3] = -0.0456; FX[26][3] = 0.4773; FX[27][3] = 4.8999; FX[28][3] = 9.7703; FX[29][3] = 7.4997; FX[30][3] = 2.1220; FX[31][3] = 0.2951; FX[32][3] = -0.2985; FX[0][4] = -0.0589; FX[1][4] = 0.5405; FX[2][4] = 5.3249; FX[3][4] = 10.1576; FX[4][4] = 6.8985; FX[5][4] = 1.8298; FX[6][4] = 0.2239; FX[7][4] = -0.4788; FX[8][4] = -3.8029; FX[9][4] = -9.3637; FX[10][4] = -8.5052; FX[11][4] = -2.6003; FX[12][4] = -0.2310; FX[13][4] = 0.1406; FX[14][4] = 2.9833; FX[15][4] = 9.0598; FX[16][4] = 9.3598; FX[17][4] = 3.2995; FX[18][4] = 0.2230; FX[19][4] = -0.0997; FX[20][4] = -1.2966; FX[21][4] = -6.7447; FX[22][4] = -10.3577; FX[23][4] = -5.5967; FX[24][4] = -0.8710; FX[25][4] = -0.0886; FX[26][4] = 0.4980; FX[27][4] = 4.8548; FX[28][4] = 10.0414; FX[29][4] = 7.4388; FX[30][4] = 2.0974; FX[31][4] = 0.1445; FX[32][4] = -0.2564; FX[0][5] = -

0.0530; FX[1][5] = 0.6558; FX[2][5] = 5.3545; FX[3][5] = 9.9671; FX[4][5] = 6.8123;
 FX[5][5] = 1.8977; FX[6][5] = 0.2658; FX[7][5] = -0.5289; FX[8][5] = -3.9398; FX[9][5]
 = -9.3002; FX[10][5] = -8.5176; FX[11][5] = -2.5442; FX[12][5] = -0.1245; FX[13][5]
 = 0.0747; FX[14][5] = 3.0192; FX[15][5] = 9.0942; FX[16][5] = 9.1832; FX[17][5] =
 3.3036; FX[18][5] = 0.3308; FX[19][5] = 0.0093; FX[20][5] = -1.3107; FX[21][5] = -
 6.6560; FX[22][5] = -10.3204; FX[23][5] = -5.6563; FX[24][5] = -1.0342; FX[25][5] =
 -0.0456; FX[26][5] = 0.4467; FX[27][5] = 4.8002; FX[28][5] = 10.3200; FX[29][5] =
 7.7279; FX[30][5] = 1.7136; FX[31][5] = 0.0627; FX[32][5] = -0.2903; FX[0][6] = -
 0.0518; FX[1][6] = 0.8970; FX[2][6] = 5.3240; FX[3][6] = 9.6191; FX[4][6] = 6.8585;
 FX[5][6] = 1.9157; FX[6][6] = 0.2935; FX[7][6] = -0.4732; FX[8][6] = -3.8263; FX[9][6]
 = -9.6346; FX[10][6] = -8.5817; FX[11][6] = -2.3848; FX[12][6] = -0.0575; FX[13][6]
 = -0.0034; FX[14][6] = 2.8259; FX[15][6] = 8.8572; FX[16][6] = 9.2485; FX[17][6] =
 3.5650; FX[18][6] = 0.4715; FX[19][6] = -0.0114; FX[20][6] = -1.3186; FX[21][6] =
 -6.7566; FX[22][6] = -10.4387; FX[23][6] = -5.4742; FX[24][6] = -0.9319; FX[25][6]
 = -0.0365; FX[26][6] = 0.4777; FX[27][6] = 5.1071; FX[28][6] = 10.4878; FX[29][6]
 = 7.5493; FX[30][6] = 1.4328; FX[31][6] = 0.0073; FX[32][6] = -0.2839; FX[0][7] =
 0.0194; FX[1][7] = 1.0222; FX[2][7] = 5.2775; FX[3][7] = 9.5759; FX[4][7] = 6.8417;
 FX[5][7] = 1.8317; FX[6][7] = 0.3475; FX[7][7] = -0.2914; FX[8][7] = -3.5830; FX[9][7]
 = -9.9998; FX[10][7] = -8.5989; FX[11][7] = -2.3523; FX[12][7] = -0.1919; FX[13][7]
 = 0.0507; FX[14][7] = 3.2783; FX[15][7] = 9.0140; FX[16][7] = 8.9090; FX[17][7] =
 3.2110; FX[18][7] = 0.3833; FX[19][7] = -0.2544; FX[20][7] = -1.6153; FX[21][7] = -
 7.0644; FX[22][7] = -9.9015; FX[23][7] = -5.1589; FX[24][7] = -0.8204; FX[25][7] =
 -0.0763; FX[26][7] = 0.6956; FX[27][7] = 5.5878; FX[28][7] = 10.5652; FX[29][7] =
 7.0645; FX[30][7] = 1.1499; FX[31][7] = 0.0147; FX[32][7] = -0.3283; FX[0][8] = -
 0.0291; FX[1][8] = 0.9846; FX[2][8] = 5.3524; FX[3][8] = 9.6238; FX[4][8] = 6.8275;
 FX[5][8] = 1.9037; FX[6][8] = 0.3117; FX[7][8] = -0.1496; FX[8][8] = -3.4857; FX[9][8]
 = -10.1389; FX[10][8] = -8.7623; FX[11][8] = -2.3023; FX[12][8] = -0.2301; FX[13][8]

= 0.2307; FX[14][8] = 3.7571; FX[15][8] = 9.3616; FX[16][8] = 8.6119; FX[17][8] = 2.7442; FX[18][8] = 0.1142; FX[19][8] = -0.3698; FX[20][8] = -2.1812; FX[21][8] = -7.5539; FX[22][8] = -9.5325; FX[23][8] = -4.4947; FX[24][8] = -0.6580; FX[25][8] = -0.0534; FX[26][8] = 0.8098; FX[27][8] = 6.0148; FX[28][8] = 10.6492; FX[29][8] = 6.5937; FX[30][8] = 0.9982; FX[31][8] = -0.0170; FX[32][8] = -0.4670; FX[0][9] = -0.0487; FX[1][9] = 0.9743; FX[2][9] = 5.5509; FX[3][9] = 9.5516; FX[4][9] = 6.8596; FX[5][9] = 1.8913; FX[6][9] = 0.2799; FX[7][9] = -0.2947; FX[8][9] = -3.9096; FX[9][9] = -10.1503; FX[10][9] = -8.4196; FX[11][9] = -2.1821; FX[12][9] = -0.1507; FX[13][9] = 0.2858; FX[14][9] = 3.9699; FX[15][9] = 9.3561; FX[16][9] = 8.4000; FX[17][9] = 2.8510; FX[18][9] = 0.1035; FX[19][9] = -0.3528; FX[20][9] = -2.5581; FX[21][9] = -8.1021; FX[22][9] = -9.4042; FX[23][9] = -4.0914; FX[24][9] = -0.4324; FX[25][9] = -0.0102; FX[26][9] = 0.9145; FX[27][9] = 6.4140; FX[28][9] = 10.7473; FX[29][9] = 6.1947; FX[30][9] = 0.8237; FX[31][9] = -0.0554; FX[32][9] = -0.3766; FX[0][10] = -0.0374; FX[1][10] = 1.3058; FX[2][10] = 5.8073; FX[3][10] = 9.5531; FX[4][10] = 6.5204; FX[5][10] = 1.6878; FX[6][10] = 0.1675; FX[7][10] = -0.4341; FX[8][10] = -4.5390; FX[9][10] = -9.9742; FX[10][10] = -7.8309; FX[11][10] = -2.0872; FX[12][10] = -0.2039; FX[13][10] = 0.3285; FX[14][10] = 3.9091; FX[15][10] = 9.2257; FX[16][10] = 8.4143; FX[17][10] = 2.8685; FX[18][10] = 0.3171; FX[19][10] = -0.4612; FX[20][10] = -2.5779; FX[21][10] = -8.0964; FX[22][10] = -9.4303; FX[23][10] = -4.1383; FX[24][10] = -0.3299; FX[25][10] = -0.0057; FX[26][10] = 1.0256; FX[27][10] = 6.9910; FX[28][10] = 10.7376; FX[29][10] = 5.5929; FX[30][10] = 0.7574; FX[31][10] = -0.0649; FX[32][10] = -0.1802; FX[0][11] = 0.0514; FX[1][11] = 1.4961; FX[2][11] = 6.1218; FX[3][11] = 9.6381; FX[4][11] = 6.0435; FX[5][11] = 1.5502; FX[6][11] = 0.1136; FX[7][11] = -0.3935; FX[8][11] = -4.6018; FX[9][11] = -9.9594; FX[10][11] = -7.7079; FX[11][11] = -2.1162; FX[12][11] = -0.2769; FX[13][11] = 0.2694; FX[14][11] = 3.9571; FX[15][11] = 9.4041; FX[16][11] = 8.2283; FX[17][11] = 2.7356; FX[18][11] = 0.4115; FX[19][11] = -0.4744; FX[20][11] = -2.5900; FX[21][11] = -7.9012; FX[22][11] = -9.4890; FX[23][11]

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= 6.2902; FX[27][24] = 10.6471; FX[28][24] = 6.1289; FX[29][24] = 1.0090; FX[30][24] = 0.0302; FX[31][24] = -0.2201; FX[32][24] = -2.4343; FX[0][25] = -0.2411; FX[1][25] = -0.9943; FX[2][25] = -3.1974; FX[3][25] = -6.4166; FX[4][25] = -6.9761; FX[5][25] = -4.5597; FX[6][25] = -1.9604; FX[7][25] = -0.2679; FX[8][25] = 0.1260; FX[9][25] = 0.8800; FX[10][25] = 2.7676; FX[11][25] = 5.0438; FX[12][25] = 6.3316; FX[13][25] = 4.6771; FX[14][25] = 2.7026; FX[15][25] = 1.3544; FX[16][25] = 0.1293; FX[17][25] = -0.5903; FX[18][25] = -1.6363; FX[19][25] = -3.8215; FX[20][25] = -7.2439; FX[21][25] = -7.3981; FX[22][25] = -3.0730; FX[23][25] = -0.2673; FX[24][25] = 0.0126; FX[25][25] = 0.9081; FX[26][25] = 6.0270; FX[27][25] = 10.3193; FX[28][25] = 6.5536; FX[29][25] = 1.1974; FX[30][25] = 0.0494; FX[31][25] = -0.2094; FX[32][25] = -2.2615; FX[0][26] = -1.0852; FX[1][26] = -3.2766; FX[2][26] = -6.4227; FX[3][26] = -7.1826; FX[4][26] = -4.5511; FX[5][26] = -1.5221; FX[6][26] = -0.3179; FX[7][26] = 0.2242; FX[8][26] = 1.4187; FX[9][26] = 3.6421; FX[10][26] = 5.2908; FX[11][26] = 5.4755; FX[12][26] = 4.2324; FX[13][26] = 2.1190; FX[14][26] = 0.7848; FX[15][26] = -0.0135; FX[16][26] = -0.5397; FX[17][26] = -0.8166; FX[18][26] = -1.4001; FX[19][26] = -3.1527; FX[20][26] = -6.6249; FX[21][26] = -6.9807; FX[22][26] = -3.1799; FX[23][26] = -0.4450; FX[24][26] = -0.0802; FX[25][26] = 0.9236; FX[26][26] = 5.7023; FX[27][26] = 10.2875; FX[28][26] = 6.8633; FX[29][26] = 1.2912; FX[30][26] = 0.0203; FX[31][26] = -0.2429; FX[32][26] = -1.8833; FX[0][27] = -3.5176; FX[1][27] = -6.1972; FX[2][27] = -6.8304; FX[3][27] = -4.8137; FX[4][27] = -1.6393; FX[5][27] = -0.2164; FX[6][27] = 0.1497; FX[7][27] = 1.7284; FX[8][27] = 4.4456; FX[9][27] = 6.3763; FX[10][27] = 5.6043; FX[11][27] = 3.2167; FX[12][27] = 1.5357; FX[13][27] = 0.2801; FX[14][27] = -0.5304; FX[15][27] = -0.9989; FX[16][27] = -1.0318; FX[17][27] = -0.7598; FX[18][27] = -0.9402; FX[19][27] = -2.2916; FX[20][27] = -5.4289; FX[21][27] = -6.4489; FX[22][27] = -3.7444; FX[23][27] = -0.9727; FX[24][27] = -0.2063; FX[25][27] = 0.8220; FX[26][27] = 5.3774; FX[27][27] = 10.0024; FX[28][27] = 7.0606; FX[29][27] = 1.6772; FX[30][27] = 0.1195; FX[31][27] = -0.1470; FX[32][27] = -1.3985; FX[0][28] = -6.7614; FX[1][28] = -6.6303; FX[2][28]

= -4.6975; FX[3][28] = -1.7280; FX[4][28] = -0.2275; FX[5][28] = 0.0450; FX[6][28] = 1.4390; FX[7][28] = 5.0796; FX[8][28] = 7.2485; FX[9][28] = 6.0009; FX[10][28] = 3.0144; FX[11][28] = 0.8424; FX[12][28] = -0.1345; FX[13][28] = -0.9869; FX[14][28] = -1.9067; FX[15][28] = -1.6507; FX[16][28] = -0.6232; FX[17][28] = 0.1243; FX[18][28] = -0.1427; FX[19][28] = -1.4831; FX[20][28] = -4.0801; FX[21][28] = -5.8852; FX[22][28] = -4.6611; FX[23][28] = -1.8831; FX[24][28] = -0.3827; FX[25][28] = 0.5983; FX[26][28] = 4.4332; FX[27][28] = 9.1334; FX[28][28] = 7.8984; FX[29][28] = 2.5617; FX[30][28] = 0.3761; FX[31][28] = 0.0181; FX[32][28] = -0.9341; FX[0][29] = -7.6586; FX[1][29] = -5.1066; FX[2][29] = -1.8194; FX[3][29] = -0.1879; FX[4][29] = 0.0897; FX[5][29] = 1.1076; FX[6][29] = 4.6168; FX[7][29] = 8.0350; FX[8][29] = 6.9666; FX[9][29] = 2.8920; FX[10][29] = 0.6505; FX[11][29] = -0.3027; FX[12][29] = -1.6265; FX[13][29] = -2.8387; FX[14][29] = -3.2690; FX[15][29] = -1.5767; FX[16][29] = 0.7125; FX[17][29] = 1.6458; FX[18][29] = 1.1489; FX[19][29] = -0.5144; FX[20][29] = -2.9529; FX[21][29] = -5.4773; FX[22][29] = -5.5310; FX[23][29] = -2.9685; FX[24][29] = -0.6932; FX[25][29] = 0.2786; FX[26][29] = 3.0608; FX[27][29] = 8.2879; FX[28][29] = 8.8485; FX[29][29] = 3.8428; FX[30][29] = 0.5809; FX[31][29] = 0.0147; FX[32][29] = -0.6278; FX[0][30] = -6.5174; FX[1][30] = -2.7344; FX[2][30] = -0.4374; FX[3][30] = 0.0520; FX[4][30] = 0.6142; FX[5][30] = 3.7955; FX[6][30] = 8.0862; FX[7][30] = 7.9667; FX[8][30] = 3.7030; FX[9][30] = 0.6197; FX[10][30] = -0.1538; FX[11][30] = -1.4855; FX[12][30] = -4.0511; FX[13][30] = -5.0585; FX[14][30] = -3.9407; FX[15][30] = -0.8272; FX[16][30] = 2.3401; FX[17][30] = 3.5955; FX[18][30] = 2.6355; FX[19][30] = 0.6987; FX[20][30] = -1.9129; FX[21][30] = -4.8092; FX[22][30] = -6.1588; FX[23][30] = -4.1675; FX[24][30] = -1.2978; FX[25][30] = 0.0382; FX[26][30] = 1.9731; FX[27][30] = 7.1692; FX[28][30] = 9.6576; FX[29][30] = 5.1642; FX[30][30] = 0.8042; FX[31][30] = 0.0188; FX[32][30] = -0.3753; FX[0][31] = -4.3253; FX[1][31] = -0.9901; FX[2][31] = -0.0691; FX[3][31] = 0.2317; FX[4][31] = 2.3265; FX[5][31] = 7.0125; FX[6][31] = 9.1047; FX[7][31] = 5.2508; FX[8][31] = 1.0530; FX[9][31] = 0.0150; FX[10][31] = -0.7881; FX[11][31] =

-3.7747; FX[12][31] = -6.9466; FX[13][31] = -6.3954; FX[14][31] = -2.8537; FX[15][31] = 0.2333; FX[16][31] = 3.5976; FX[17][31] = 5.4345; FX[18][31] = 4.1883; FX[19][31] = 1.7901; FX[20][31] = -1.0023; FX[21][31] = -3.8799; FX[22][31] = -6.6816; FX[23][31] = -5.4040; FX[24][31] = -2.1905; FX[25][31] = -0.2317; FX[26][31] = 1.0881; FX[27][31] = 5.7884; FX[28][31] = 10.0942; FX[29][31] = 6.4867; FX[30][31] = 1.4007; FX[31][31] = 0.0620; FX[32][31] = -0.0842; FX[0][32] = -1.8741; FX[1][32] = -0.1488; FX[2][32] = 0.0075; FX[3][32] = 0.9838; FX[4][32] = 4.8009; FX[5][32] = 8.9075; FX[6][32] = 7.5691; FX[7][32] = 2.6450; FX[8][32] = 0.1428; FX[9][32] = -0.2313; FX[10][32] = -1.8637; FX[11][32] = -6.6991; FX[12][32] = -8.8524; FX[13][32] = -5.2042; FX[14][32] = -1.2429; FX[15][32] = 0.7759; FX[16][32] = 4.1161; FX[17][32] = 7.0914; FX[18][32] = 5.7766; FX[19][32] = 2.4261; FX[20][32] = -0.3903; FX[21][32] = -3.5318; FX[22][32] = -6.8545; FX[23][32] = -6.7515; FX[24][32] = -3.0976; FX[25][32] = -0.5105; FX[26][32] = 0.5737; FX[27][32] = 4.6623; FX[28][32] = 9.8355; FX[29][32] = 7.7467; FX[30][32] = 1.9759; FX[31][32] = 0.1758; FX[32][32] = -0.0419; FX[0][33] = -0.6376; FX[1][33] = -0.0031; FX[2][33] = 0.1163; FX[3][33] = 2.3501; FX[4][33] = 7.1662; FX[5][33] = 8.9435; FX[6][33] = 5.2084; FX[7][33] = 1.1966; FX[8][33] = 0.0101; FX[9][33] = -0.5165; FX[10][33] = -3.8498; FX[11][33] = -9.0092; FX[12][33] = -8.2109; FX[13][33] = -2.9888; FX[14][33] = -0.3955; FX[15][33] = 0.8652; FX[16][33] = 4.2833; FX[17][33] = 8.0968; FX[18][33] = 7.0656; FX[19][33] = 2.5337; FX[20][33] = -0.2613; FX[21][33] = -3.1825; FX[22][33] = -7.0373; FX[23][33] = -7.7333; FX[24][33] = -4.0158; FX[25][33] = -0.5727; FX[26][33] = 0.5829; FX[27][33] = 3.8882; FX[28][33] = 9.1786; FX[29][33] = 8.4053; FX[30][33] = 2.7201; FX[31][33] = 0.1635; FX[32][33] = -0.0752; FX[0][34] = -0.2549; FX[1][34] = -0.0917; FX[2][34] = 0.6675; FX[3][34] = 4.2413; FX[4][34] = 8.5575; FX[5][34] = 7.7721; FX[6][34] = 3.2345; FX[7][34] = 0.5904; FX[8][34] = -0.0388; FX[9][34] = -1.2691; FX[10][34] = -6.2667; FX[11][34] = -9.9690; FX[12][34] = -6.0927; FX[13][34] = -1.3481; FX[14][34] = -0.0930; FX[15][34] = 0.8561; FX[16][34] = 5.1306; FX[17][34] = 9.2978; FX[18][34] = 6.9082; FX[19][34] = 1.9396; FX[20][34] =

-0.3644; FX[21][34] = -2.6907; FX[22][34] = -7.1982; FX[23][34] = -8.5600; FX[24][34] = -4.5244; FX[25][34] = -0.6743; FX[26][34] = 0.5719; FX[27][34] = 3.3257; FX[28][34] = 8.5855; FX[29][34] = 8.8285; FX[30][34] = 3.4039; FX[31][34] = 0.2364; FX[32][34] = -0.0604; FX[0][35] = -0.0921; FX[1][35] = -0.0174; FX[2][35] = 1.2848; FX[3][35] = 5.7258; FX[4][35] = 9.2458; FX[5][35] = 6.5447; FX[6][35] = 1.9623; FX[7][35] = 0.2688; FX[8][35] = -0.0343; FX[9][35] = -2.3853; FX[10][35] = -8.5706; FX[11][35] = -9.7715; FX[12][35] = -3.8510; FX[13][35] = -0.4568; FX[14][35] = 0.0071; FX[15][35] = 1.1311; FX[16][35] = 6.3875; FX[17][35] = 10.1748; FX[18][35] = 6.0690; FX[19][35] = 0.9902; FX[20][35] = -0.4093; FX[21][35] = -2.4377; FX[22][35] = -7.3463; FX[23][35] = -9.0535; FX[24][35] = -4.7125; FX[25][35] = -0.7721; FX[26][35] = 0.3943; FX[27][35] = 2.9192; FX[28][35] = 8.4223; FX[29][35] = 9.2713; FX[30][35] = 3.6175; FX[31][35] = 0.3811; FX[32][35] = -0.0363; FX[0][36] = -0.0345; FX[1][36] = 0.1862; FX[2][36] = 1.8269; FX[3][36] = 6.8144; FX[4][36] = 9.5885; FX[5][36] = 5.3778; FX[6][36] = 1.1615; FX[7][36] = 0.1050; FX[8][36] = -0.3038; FX[9][36] = -4.1248; FX[10][36] = -10.2519; FX[11][36] = -8.2659; FX[12][36] = -2.0943; FX[13][36] = -0.0272; FX[14][36] = 0.0542; FX[15][36] = 1.5977; FX[16][36] = 7.4097; FX[17][36] = 10.2719; FX[18][36] = 5.1635; FX[19][36] = 0.4881; FX[20][36] = -0.4482; FX[21][36] = -2.7361; FX[22][36] = -7.7974; FX[23][36] = -9.0893; FX[24][36] = -4.1834; FX[25][36] = -0.7383; FX[26][36] = 0.2570; FX[27][36] = 2.6143; FX[28][36] = 8.5300; FX[29][36] = 9.4022; FX[30][36] = 3.9124; FX[31][36] = 0.3280; FX[32][36] = -0.0566; FX[0][37] = -0.0486; FX[1][37] = 0.4175; FX[2][37] = 2.5255; FX[3][37] = 7.8073; FX[4][37] = 9.3853; FX[5][37] = 4.2599; FX[6][37] = 0.6496; FX[7][37] = 0.0180; FX[8][37] = -0.8184; FX[9][37] = -6.2981; FX[10][37] = -10.8718; FX[11][37] = -6.3375; FX[12][37] = -0.7903; FX[13][37] = 0.0538; FX[14][37] = 0.1419; FX[15][37] = 2.5230; FX[16][37] = 8.5253; FX[17][37] = 9.6460; FX[18][37] = 3.9976; FX[19][37] = 0.2176; FX[20][37] = -0.3778; FX[21][37] = -2.9691; FX[22][37] = -8.3799; FX[23][37] = -9.0431; FX[24][37] = -3.8060; FX[25][37] = -0.4856; FX[26][37] = 0.2410; FX[27][37] = 2.6348; FX[28][37] = 8.5551; FX[29][37]

= 9.4866; FX[30][37] = 3.8786; FX[31][37] = 0.2599; FX[32][37] = -0.0879; FX[0][38] = -0.0917; FX[1][38] = 0.5613; FX[2][38] = 3.2318; FX[3][38] = 8.6528; FX[4][38] = 8.8535; FX[5][38] = 3.3514; FX[6][38] = 0.4092; FX[7][38] = -0.0189; FX[8][38] = -1.2121; FX[9][38] = -7.4460; FX[10][38] = -10.8432; FX[11][38] = -5.2074; FX[12][38] = -0.3643; FX[13][38] = 0.0198; FX[14][38] = 0.2937; FX[15][38] = 3.8543; FX[16][38] = 9.7992; FX[17][38] = 8.4954; FX[18][38] = 2.5760; FX[19][38] = 0.0379; FX[20][38] = -0.3550; FX[21][38] = -2.9889; FX[22][38] = -8.6590; FX[23][38] = -9.2604; FX[24][38] = -3.4398; FX[25][38] = -0.3727; FX[26][38] = 0.2405; FX[27][38] = 3.0164; FX[28][38] = 8.8127; FX[29][38] = 9.2114; FX[30][38] = 3.5385; FX[31][38] = 0.2665; FX[32][38] = -0.1575; FX[0][39] = -0.0629; FX[1][39] = 0.5453; FX[2][39] = 3.8424; FX[3][39] = 9.1481; FX[4][39] = 8.2271; FX[5][39] = 2.8468; FX[6][39] = 0.4049; FX[7][39] = -0.1114; FX[8][39] = -1.4788; FX[9][39] = -7.6630; FX[10][39] = -10.5595; FX[11][39] = -4.8261; FX[12][39] = -0.3958; FX[13][39] = -0.0119; FX[14][39] = 0.6566; FX[15][39] = 5.1684; FX[16][39] = 10.2329; FX[17][39] = 7.4145; FX[18][39] = 1.5683; FX[19][39] = -0.0476; FX[20][39] = -0.4214; FX[21][39] = -3.4655; FX[22][39] = -9.0551; FX[23][39] = -8.8173; FX[24][39] = -2.9713; FX[25][39] = -0.2683; FX[26][39] = 0.2688; FX[27][39] = 2.9683; FX[28][39] = 8.8947; FX[29][39] = 9.3507; FX[30][39] = 3.3303; FX[31][39] = 0.2621; FX[32][39] = -0.1309; FX[0][40] = -0.0307; FX[1][40] = 0.5671; FX[2][40] = 4.3840; FX[3][40] = 9.2250; FX[4][40] = 7.7217; FX[5][40] = 2.6667; FX[6][40] = 0.3948; FX[7][40] = -0.1867; FX[8][40] = -1.9093; FX[9][40] = -7.7369; FX[10][40] = -10.1387; FX[11][40] = -4.6647; FX[12][40] = -0.3920; FX[13][40] = 0.0020; FX[14][40] = 1.1158; FX[15][40] = 6.1942; FX[16][40] = 10.2419; FX[17][40] = 6.2886; FX[18][40] = 1.1042; FX[19][40] = -0.0164; FX[20][40] = -0.4146; FX[21][40] = -4.0974; FX[22][40] = -9.6591; FX[23][40] = -8.2471; FX[24][40] = -2.3864; FX[25][40] = -0.1313; FX[26][40] = 0.3335; FX[27][40] = 3.0610; FX[28][40] = 8.7039; FX[29][40] = 9.3152; FX[30][40] = 3.4162; FX[31][40] = 0.2043; FX[32][40] = -0.0916; FX[0][41] = -0.0180; FX[1][41] = 0.7759; FX[2][41] = 4.6742; FX[3][41] = 9.4092; FX[4][41] = 7.4568; FX[5][41] =

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 V[2][3] = 0.7762; V[3][3] = 0.1032; V[4][3] = -0.6279; V[5][3] = -0.9409; V[6][3] =
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$V[11][3] = 0.9364$; $V[12][3] = 1.0020$; $V[13][3] = 1.0035$; $V[14][3] = 0.9284$; $V[15][3] = 0.4790$; $V[16][3] = -0.3240$; $V[17][3] = -0.8743$; $V[18][3] = -0.9992$; $V[19][3] = -0.9991$; $V[20][3] = -0.9562$; $V[21][3] = -0.6617$; $V[22][3] = 0.0795$; $V[23][3] = 0.7743$; $V[24][3] = 0.9857$; $V[25][3] = 1.0009$; $V[26][3] = 0.9967$; $V[27][3] = 0.8181$; $V[28][3] = 0.1982$; $V[29][3] = -0.5555$; $V[30][3] = -0.9187$; $V[31][3] = -0.9969$; $V[32][3] = -0.9925$; $V[0][4] = 1.0009$; $V[1][4] = 0.9979$; $V[2][4] = 0.7979$; $V[3][4] = 0.1393$; $V[4][4] = -0.6034$; $V[5][4] = -0.9262$; $V[6][4] = -0.9887$; $V[7][4] = -0.9847$; $V[8][4] = -0.8482$; $V[9][4] = -0.3018$; $V[10][4] = 0.4748$; $V[11][4] = 0.9182$; $V[12][4] = 0.9992$; $V[13][4] = 1.0048$; $V[14][4] = 0.9134$; $V[15][4] = 0.4299$; $V[16][4] = -0.3787$; $V[17][4] = -0.8941$; $V[18][4] = -0.9982$; $V[19][4] = -0.9983$; $V[20][4] = -0.9647$; $V[21][4] = -0.6683$; $V[22][4] = 0.0687$; $V[23][4] = 0.7564$; $V[24][4] = 0.9767$; $V[25][4] = 1.0005$; $V[26][4] = 0.9960$; $V[27][4] = 0.8199$; $V[28][4] = 0.1890$; $V[29][4] = -0.5718$; $V[30][4] = -0.9348$; $V[31][4] = -1.0017$; $V[32][4] = -0.9942$; $V[0][5] = 1.0013$; $V[1][5] = 0.9922$; $V[2][5] = 0.7861$; $V[3][5] = 0.1336$; $V[4][5] = -0.5956$; $V[5][5] = -0.9184$; $V[6][5] = -0.9868$; $V[7][5] = -0.9824$; $V[8][5] = -0.8360$; $V[9][5] = -0.2902$; $V[10][5] = 0.4881$; $V[11][5] = 0.9281$; $V[12][5] = 1.0020$; $V[13][5] = 1.0045$; $V[14][5] = 0.9187$; $V[15][5] = 0.4287$; $V[16][5] = -0.3747$; $V[17][5] = -0.8786$; $V[18][5] = -0.9908$; $V[19][5] = -0.9992$; $V[20][5] = -0.9680$; $V[21][5] = -0.6779$; $V[22][5] = 0.0578$; $V[23][5] = 0.7423$; $V[24][5] = 0.9760$; $V[25][5] = 1.0027$; $V[26][5] = 0.9981$; $V[27][5] = 0.8275$; $V[28][5] = 0.1894$; $V[29][5] = -0.6009$; $V[30][5] = -0.9570$; $V[31][5] = -1.0017$; $V[32][5] = -0.9926$; $V[0][6] = 0.9983$; $V[1][6] = 0.9788$; $V[2][6] = 0.7611$; $V[3][6] = 0.1243$; $V[4][6] = -0.5853$; $V[5][6] = -0.9182$; $V[6][6] = -0.9868$; $V[7][6] = -0.9839$; $V[8][6] = -0.8463$; $V[9][6] = -0.2957$; $V[10][6] = 0.5096$; $V[11][6] = 0.9379$; $V[12][6] = 1.0038$; $V[13][6] = 1.0050$; $V[14][6] = 0.9337$; $V[15][6] = 0.4583$; $V[16][6] = -0.3344$; $V[17][6] = -0.8525$; $V[18][6] = -0.9852$; $V[19][6] = -0.9951$; $V[20][6] = -0.9635$; $V[21][6] = -0.6701$; $V[22][6] = 0.0776$; $V[23][6] = 0.7589$; $V[24][6] = 0.9782$; $V[25][6] = 1.0026$; $V[26][6] = 0.9965$; $V[27][6] = 0.8143$; $V[28][6] = 0.1502$; $V[29][6] = -0.6351$; $V[30][6] = -0.9729$; $V[31][6] = -1.0022$; $V[32][6] = -0.9935$;

$V[0][7] = 0.9940$; $V[1][7] = 0.9657$; $V[2][7] = 0.7439$; $V[3][7] = 0.1132$; $V[4][7] = -$
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 $V[15][9] = 0.3162$; $V[16][9] = -0.4587$; $V[17][9] = -0.9077$; $V[18][9] = -0.9948$; $V[19][9]$
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-0.8893; V[21][10] = -0.4739; V[22][10] = 0.2911; V[23][10] = 0.8572; V[24][10] =
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 0.0753; V[29][10] = -0.7745; V[30][10] = -0.9916; V[31][10] = -1.0022; V[32][10] =
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 = -0.8351; V[9][11] = -0.2194; V[10][11] = 0.5506; V[11][11] = 0.9260; V[12][11] =
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 0.4549; V[17][11] = -0.8833; V[18][11] = -0.9882; V[19][11] = -0.9838; V[20][11] =
 -0.8848; V[21][11] = -0.4723; V[22][11] = 0.2782; V[23][11] = 0.8585; V[24][11] =
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 0.1132; V[29][11] = -0.7916; V[30][11] = -0.9894; V[31][11] = -1.0013; V[32][11] =
 -0.9907; V[0][12] = 1.0015; V[1][12] = 0.9600; V[2][12] = 0.6602; V[3][12] = -0.0225;
 V[4][12] = -0.6872; V[5][12] = -0.9535; V[6][12] = -0.9988; V[7][12] = -0.9974; V[8][12]
 = -0.8596; V[9][12] = -0.2576; V[10][12] = 0.5375; V[11][12] = 0.9328; V[12][12] =
 0.9994; V[13][12] = 1.0018; V[14][12] = 0.8709; V[15][12] = 0.3086; V[16][12] = -
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 = -0.8754; V[9][13] = -0.2898; V[10][13] = 0.5251; V[11][13] = 0.9396; V[12][13] =
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 $V[21][32] = -0.5439$; $V[22][32] = -0.1226$; $V[23][32] = 0.4621$; $V[24][32] = 0.8660$;
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= 0.9654; V[14][34] = 1.0052; V[15][34] = 0.9887; V[16][34] = 0.7761; V[17][34] = 0.1653; V[18][34] = -0.5336; V[19][34] = -0.8729; V[20][34] = -0.9216; V[21][34] = -0.8147; V[22][34] = -0.4245; V[23][34] = 0.2518; V[24][34] = 0.8021; V[25][34] = 0.9846; V[26][34] = 0.9825; V[27][34] = 0.8531; V[28][34] = 0.3732; V[29][34] = -0.3831; V[30][34] = -0.8873; V[31][34] = -0.9959; V[32][34] = -0.9979; V[0][35] = 0.9969; V[1][35] = 1.0022; V[2][35] = 0.9711; V[3][35] = 0.7111; V[4][35] = 0.0740; V[5][35] = -0.5985; V[6][35] = -0.9257; V[7][35] = -0.9945; V[8][35] = -1.0023; V[9][35] = -0.9416; V[10][35] = -0.5061; V[11][35] = 0.2973; V[12][35] = 0.8573; V[13][35] = 0.9966; V[14][35] = 1.0040; V[15][35] = 0.9804; V[16][35] = 0.7087; V[17][35] = -0.0079; V[18][35] = -0.7037; V[19][35] = -0.9569; V[20][35] = -0.9668; V[21][35] = -0.8736; V[22][35] = -0.4926; V[23][35] = 0.2168; V[24][35] = 0.7931; V[25][35] = 0.9868; V[26][35] = 0.9936; V[27][35] = 0.8878; V[28][35] = 0.4382; V[29][35] = -0.3363; V[30][35] = -0.8654; V[31][35] = -0.9910; V[32][35] = -0.9974; V[0][36] = 1.0007; V[1][36] = 0.9982; V[2][36] = 0.9394; V[3][36] = 0.6106; V[4][36] = -0.0973; V[5][36] = -0.7280; V[6][36] = -0.9670; V[7][36] = -0.9997; V[8][36] = -0.9988; V[9][36] = -0.8659; V[10][36] = -0.2611; V[11][36] = 0.5484; V[12][36] = 0.9507; V[13][36] = 1.0035; V[14][36] = 1.0020; V[15][36] = 0.9609; V[16][36] = 0.6248; V[17][36] = -0.1477; V[18][36] = -0.8021; V[19][36] = -0.9907; V[20][36] = -0.9856; V[21][36] = -0.8807; V[22][36] = -0.4670; V[23][36] = 0.2670; V[24][36] = 0.8178; V[25][36] = 0.9854; V[26][36] = 0.9991; V[27][36] = 0.9133; V[28][36] = 0.4712; V[29][36] = -0.3113; V[30][36] = -0.8622; V[31][36] = -0.9964; V[32][36] = -0.9983; V[0][37] = 1.0007; V[1][37] = 0.9902; V[2][37] = 0.8987; V[3][37] = 0.4891; V[4][37] = -0.2509; V[5][37] = -0.8253; V[6][37] = -0.9877; V[7][37] = -1.0034; V[8][37] = -0.9893; V[9][37] = -0.7443; V[10][37] = 0.0007; V[11][37] = 0.7458; V[12][37] = 0.9932; V[13][37] = 1.0028; V[14][37] = 0.9993; V[15][37] = 0.9255; V[16][37] = 0.4870; V[17][37] = -0.3040; V[18][37] = -0.8747; V[19][37] = -1.0013; V[20][37] = -0.9961; V[21][37] = -0.8858; V[22][37] = -0.4384; V[23][37] = 0.3240; V[24][37] = 0.8539; V[25][37] =

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 -0.8899 ; $V[22][38] = -0.4273$; $V[23][38] = 0.3594$; $V[24][38] = 0.8783$; $V[25][38] =$
 0.9948 ; $V[26][38] = 1.0009$; $V[27][38] = 0.9013$; $V[28][38] = 0.4270$; $V[29][38] = -$
 0.3623 ; $V[30][38] = -0.8820$; $V[31][38] = -1.0014$; $V[32][38] = -0.9981$; $V[0][39] =$
 0.9996 ; $V[1][39] = 0.9878$; $V[2][39] = 0.8446$; $V[3][39] = 0.3077$; $V[4][39] = -0.4457$;
 $V[5][39] = -0.8900$; $V[6][39] = -0.9912$; $V[7][39] = -0.9985$; $V[8][39] = -0.9586$; $V[9][39]$
 $= -0.6225$; $V[10][39] = 0.1809$; $V[11][39] = 0.8293$; $V[12][39] = 0.9983$; $V[13][39] =$
 1.0021 ; $V[14][39] = 0.9905$; $V[15][39] = 0.7934$; $V[16][39] = 0.1432$; $V[17][39] = -$
 0.6295 ; $V[18][39] = -0.9660$; $V[19][39] = -0.9995$; $V[20][39] = -0.9867$; $V[21][39] =$
 -0.8666 ; $V[22][39] = -0.3525$; $V[23][39] = 0.4255$; $V[24][39] = 0.9020$; $V[25][39] =$
 0.9984 ; $V[26][39] = 0.9983$; $V[27][39] = 0.9016$; $V[28][39] = 0.4263$; $V[29][39] = -$
 0.3743 ; $V[30][39] = -0.8910$; $V[31][39] = -0.9994$; $V[32][39] = -0.9977$; $V[0][40] =$
 0.9971 ; $V[1][40] = 0.9852$; $V[2][40] = 0.8201$; $V[3][40] = 0.2495$; $V[4][40] = -0.4817$;
 $V[5][40] = -0.8937$; $V[6][40] = -0.9903$; $V[7][40] = -0.9964$; $V[8][40] = -0.9371$; $V[9][40]$
 $= -0.5708$; $V[10][40] = 0.2108$; $V[11][40] = 0.8342$; $V[12][40] = 0.9980$; $V[13][40] =$
 1.0032 ; $V[14][40] = 0.9766$; $V[15][40] = 0.7166$; $V[16][40] = 0.0078$; $V[17][40] = -$
 0.7042 ; $V[18][40] = -0.9704$; $V[19][40] = -0.9933$; $V[20][40] = -0.9849$; $V[21][40] =$
 -0.8422 ; $V[22][40] = -0.2676$; $V[23][40] = 0.5125$; $V[24][40] = 0.9307$; $V[25][40] =$
 1.0022 ; $V[26][40] = 0.9949$; $V[27][40] = 0.8923$; $V[28][40] = 0.4178$; $V[29][40] = -$
 0.3674 ; $V[30][40] = -0.8911$; $V[31][40] = -0.9985$; $V[32][40] = -0.9967$; $V[0][41] =$
 0.9981 ; $V[1][41] = 0.9779$; $V[2][41] = 0.7930$; $V[3][41] = 0.1990$; $V[4][41] = -0.5297$;

$V[5][41] = -0.9118$; $V[6][41] = -0.9952$; $V[7][41] = -0.9957$; $V[8][41] = -0.9118$; $V[9][41]$
 $= -0.5094$; $V[10][41] = 0.2567$; $V[11][41] = 0.8397$; $V[12][41] = 1.0009$; $V[13][41] =$
 1.0037 ; $V[14][41] = 0.9730$; $V[15][41] = 0.6649$; $V[16][41] = -0.0783$; $V[17][41] = -$
 0.7528 ; $V[18][41] = -0.9804$; $V[19][41] = -0.9941$; $V[20][41] = -0.9840$; $V[21][41] =$
 -0.7999 ; $V[22][41] = -0.1788$; $V[23][41] = 0.5628$; $V[24][41] = 0.9407$; $V[25][41] =$
 1.0011 ; $V[26][41] = 0.9935$; $V[27][41] = 0.8693$; $V[28][41] = 0.3766$; $V[29][41] = -$
 0.4016 ; $V[30][41] = -0.8908$; $V[31][41] = -0.9974$; $V[32][41] = -0.9975$; $V[0][42] =$
 1.0001 ; $V[1][42] = 0.9745$; $V[2][42] = 0.7736$; $V[3][42] = 0.1490$; $V[4][42] = -0.5831$;
 $V[5][42] = -0.9356$; $V[6][42] = -0.9911$; $V[7][42] = -0.9843$; $V[8][42] = -0.8778$; $V[9][42]$
 $= -0.4407$; $V[10][42] = 0.3067$; $V[11][42] = 0.8419$; $V[12][42] = 0.9985$; $V[13][42] =$
 1.0039 ; $V[14][42] = 0.9758$; $V[15][42] = 0.6554$; $V[16][42] = -0.1274$; $V[17][42] = -$
 0.7973 ; $V[18][42] = -0.9971$; $V[19][42] = -1.0007$; $V[20][42] = -0.9733$; $V[21][42] =$
 -0.7446 ; $V[22][42] = -0.1041$; $V[23][42] = 0.6000$; $V[24][42] = 0.9375$; $V[25][42] =$
 0.9994 ; $V[26][42] = 1.0013$; $V[27][42] = 0.8728$; $V[28][42] = 0.3470$; $V[29][42] = -$
 0.4384 ; $V[30][42] = -0.9062$; $V[31][42] = -0.9982$; $V[32][42] = -0.9976$; $V[0][43] =$
 1.0007 ; $V[1][43] = 0.9728$; $V[2][43] = 0.7559$; $V[3][43] = 0.1380$; $V[4][43] = -0.5905$;
 $V[5][43] = -0.9292$; $V[6][43] = -0.9900$; $V[7][43] = -0.9798$; $V[8][43] = -0.8607$; $V[9][43]$
 $= -0.4156$; $V[10][43] = 0.3205$; $V[11][43] = 0.8472$; $V[12][43] = 0.9967$; $V[13][43] =$
 1.0049 ; $V[14][43] = 0.9689$; $V[15][43] = 0.6419$; $V[16][43] = -0.1387$; $V[17][43] = -$
 0.8098 ; $V[18][43] = -0.9972$; $V[19][43] = -1.0003$; $V[20][43] = -0.9634$; $V[21][43] =$
 -0.7013 ; $V[22][43] = -0.0419$; $V[23][43] = 0.6395$; $V[24][43] = 0.9396$; $V[25][43] =$
 0.9995 ; $V[26][43] = 1.0009$; $V[27][43] = 0.8782$; $V[28][43] = 0.3383$; $V[29][43] = -$
 0.4385 ; $V[30][43] = -0.9085$; $V[31][43] = -1.0004$; $V[32][43] = -0.9990$; $V[0][44] =$
 1.0005 ; $V[1][44] = 0.9791$; $V[2][44] = 0.7901$; $V[3][44] = 0.1805$; $V[4][44] = -0.5571$;
 $V[5][44] = -0.9276$; $V[6][44] = -0.9959$; $V[7][44] = -0.9871$; $V[8][44] = -0.8914$; $V[9][44]$
 $= -0.4691$; $V[10][44] = 0.2844$; $V[11][44] = 0.8363$; $V[12][44] = 0.9944$; $V[13][44] =$
 1.0051 ; $V[14][44] = 0.9564$; $V[15][44] = 0.6241$; $V[16][44] = -0.1383$; $V[17][44] = -$

0.7883; $V[18][44] = -0.9903$; $V[19][44] = -0.9973$; $V[20][44] = -0.9504$; $V[21][44] = -0.6741$; $V[22][44] = -0.0157$; $V[23][44] = 0.6587$; $V[24][44] = 0.9530$; $V[25][44] = 1.0008$; $V[26][44] = 0.9980$; $V[27][44] = 0.8516$; $V[28][44] = 0.3101$; $V[29][44] = -0.4350$; $V[30][44] = -0.8916$; $V[31][44] = -0.9996$; $V[32][44] = -0.9988$; $V[0][45] = 0.9996$; $V[1][45] = 0.9936$; $V[2][45] = 0.8477$; $V[3][45] = 0.2655$; $V[4][45] = -0.5081$; $V[5][45] = -0.9233$; $V[6][45] = -0.9932$; $V[7][45] = -0.9929$; $V[8][45] = -0.9269$; $V[9][45] = -0.5366$; $V[10][45] = 0.2273$; $V[11][45] = 0.8241$; $V[12][45] = 0.9955$; $V[13][45] = 1.0062$; $V[14][45] = 0.9586$; $V[15][45] = 0.6266$; $V[16][45] = -0.1152$; $V[17][45] = -0.7682$; $V[18][45] = -0.9869$; $V[19][45] = -0.9978$; $V[20][45] = -0.9423$; $V[21][45] = -0.6804$; $V[22][45] = -0.0434$; $V[23][45] = 0.6421$; $V[24][45] = 0.9611$; $V[25][45] = 1.0039$; $V[26][45] = 0.9965$; $V[27][45] = 0.8391$; $V[28][45] = 0.2880$; $V[29][45] = -0.4496$; $V[30][45] = -0.8940$; $V[31][45] = -1.0007$; $V[32][45] = -0.9976$; $V[0][46] = 0.9999$; $V[1][46] = 0.9999$; $V[2][46] = 0.8643$; $V[3][46] = 0.3140$; $V[4][46] = -0.4621$; $V[5][46] = -0.9011$; $V[6][46] = -0.9883$; $V[7][46] = -0.9954$; $V[8][46] = -0.9467$; $V[9][46] = -0.5994$; $V[10][46] = 0.1690$; $V[11][46] = 0.8056$; $V[12][46] = 0.9955$; $V[13][46] = 1.0045$; $V[14][46] = 0.9706$; $V[15][46] = 0.6557$; $V[16][46] = -0.0924$; $V[17][46] = -0.7668$; $V[18][46] = -0.9904$; $V[19][46] = -0.9988$; $V[20][46] = -0.9556$; $V[21][46] = -0.7041$; $V[22][46] = -0.0601$; $V[23][46] = 0.6521$; $V[24][46] = 0.9716$; $V[25][46] = 1.0022$; $V[26][46] = 0.9960$; $V[27][46] = 0.8292$; $V[28][46] = 0.2579$; $V[29][46] = -0.4923$; $V[30][46] = -0.9150$; $V[31][46] = -1.0032$; $V[32][46] = -0.9994$; $V[0][47] = 0.9994$; $V[1][47] = 0.9991$; $V[2][47] = 0.8726$; $V[3][47] = 0.3627$; $V[4][47] = -0.3906$; $V[5][47] = -0.8671$; $V[6][47] = -0.9860$; $V[7][47] = -0.9978$; $V[8][47] = -0.9593$; $V[9][47] = -0.6301$; $V[10][47] = 0.1269$; $V[11][47] = 0.7812$; $V[12][47] = 0.9946$; $V[13][47] = 1.0034$; $V[14][47] = 0.9643$; $V[15][47] = 0.6611$; $V[16][47] = -0.0791$; $V[17][47] = -0.7592$; $V[18][47] = -0.9873$; $V[19][47] = -0.9926$; $V[20][47] = -0.9576$; $V[21][47] = -0.7317$; $V[22][47] = -0.0721$; $V[23][47] = 0.6613$; $V[24][47] = 0.9751$; $V[25][47] = 1.0020$; $V[26][47] = 0.9923$; $V[27][47] = 0.8345$; $V[28][47] = 0.2710$; $V[29][47] = -$

0.5006; $V[30][47] = -0.9245$; $V[31][47] = -1.0020$; $V[32][47] = -0.9994$; $V[0][48] = 0.9986$; $V[1][48] = 1.0032$; $V[2][48] = 0.9033$; $V[3][48] = 0.4375$; $V[4][48] = -0.3088$; $V[5][48] = -0.8254$; $V[6][48] = -0.9792$; $V[7][48] = -1.0000$; $V[8][48] = -0.9616$; $V[9][48] = -0.6156$; $V[10][48] = 0.1543$; $V[11][48] = 0.8090$; $V[12][48] = 0.9971$; $V[13][48] = 1.0033$; $V[14][48] = 0.9539$; $V[15][48] = 0.6310$; $V[16][48] = -0.0834$; $V[17][48] = -0.7336$; $V[18][48] = -0.9705$; $V[19][48] = -0.9909$; $V[20][48] = -0.9550$; $V[21][48] = -0.7391$; $V[22][48] = -0.1011$; $V[23][48] = 0.6355$; $V[24][48] = 0.9678$; $V[25][48] = 1.0040$; $V[26][48] = 0.9904$; $V[27][48] = 0.8498$; $V[28][48] = 0.2785$; $V[29][48] = -0.5099$; $V[30][48] = -0.9339$; $V[31][48] = -0.9993$; $V[32][48] = -0.9991$; $V[0][49] = 0.9945$; $V[1][49] = 1.0045$; $V[2][49] = 0.9198$; $V[3][49] = 0.4471$; $V[4][49] = -0.3036$; $V[5][49] = -0.8054$; $V[6][49] = -0.9695$; $V[7][49] = -0.9998$; $V[8][49] = -0.9646$; $V[9][49] = -0.6214$; $V[10][49] = 0.1486$; $V[11][49] = 0.8119$; $V[12][49] = 0.9990$; $V[13][49] = 1.0033$; $V[14][49] = 0.9557$; $V[15][49] = 0.6401$; $V[16][49] = -0.0570$; $V[17][49] = -0.7034$; $V[18][49] = -0.9569$; $V[19][49] = -0.9959$; $V[20][49] = -0.9732$; $V[21][49] = -0.7684$; $V[22][49] = -0.1184$; $V[23][49] = 0.6343$; $V[24][49] = 0.9704$; $V[25][49] = 1.0039$; $V[26][49] = 0.9980$; $V[27][49] = 0.8450$; $V[28][49] = 0.2506$; $V[29][49] = -0.5482$; $V[30][49] = -0.9453$; $V[31][49] = -0.9995$; $V[32][49] = -1.0000$; $V[0][50] = 0.9938$; $V[1][50] = 1.0031$; $V[2][50] = 0.9235$; $V[3][50] = 0.4417$; $V[4][50] = -0.3105$; $V[5][50] = -0.7998$; $V[6][50] = -0.9550$; $V[7][50] = -0.9975$; $V[8][50] = -0.9643$; $V[9][50] = -0.6746$; $V[10][50] = 0.0737$; $V[11][50] = 0.7671$; $V[12][50] = 0.9957$; $V[13][50] = 1.0033$; $V[14][50] = 0.9720$; $V[15][50] = 0.6927$; $V[16][50] = 0.0190$; $V[17][50] = -0.6454$; $V[18][50] = -0.9444$; $V[19][50] = -0.9952$; $V[20][50] = -0.9755$; $V[21][50] = -0.7646$; $V[22][50] = -0.0936$; $V[23][50] = 0.6609$; $V[24][50] = 0.9750$; $V[25][50] = 1.0033$; $V[26][50] = 0.9993$; $V[27][50] = 0.8485$; $V[28][50] = 0.2495$; $V[29][50] = -0.5487$; $V[30][50] = -0.9466$; $V[31][50] = -1.0017$; $V[32][50] = -0.9994$; $V[0][51] = 0.9941$; $V[1][51] = 1.0034$; $V[2][51] = 0.9255$; $V[3][51] = 0.4851$; $V[4][51] = -0.2393$; $V[5][51] = -0.7540$; $V[6][51] = -0.9395$; $V[7][51] = -0.9943$; $V[8][51] = -0.9680$; $V[9][51]$

= -0.7120; V[10][51] = 0.0047; V[11][51] = 0.7313; V[12][51] = 0.9941; V[13][51] = 1.0029; V[14][51] = 0.9775; V[15][51] = 0.7333; V[16][51] = 0.0685; V[17][51] = -0.6269; V[18][51] = -0.9346; V[19][51] = -0.9887; V[20][51] = -0.9660; V[21][51] = -0.7450; V[22][51] = -0.0755; V[23][51] = 0.6727; V[24][51] = 0.9683; V[25][51] = 1.0035; V[26][51] = 0.9974; V[27][51] = 0.8348; V[28][51] = 0.2320; V[29][51] = -0.5517; V[30][51] = -0.9444; V[31][51] = -1.0026; V[32][51] = -0.9994;

B.0.4 Defect Order = 4

$if(m_{index} == 1 \&\& (m_{pos.x} - 0.25 * pitch) > (-2.75 * pitch) \&\& (m_{pos.x} - 0.25 * pitch) < (2.75 * pitch) \&\& m_{pos.y} > (-2.0 * pitch) \&\& m_{pos.y} < (2.0 * pitch))$

FX[0][0] = 9.9609; FX[1][0] = 5.0987; FX[2][0] = 0.7534; FX[3][0] = -0.1177; FX[4][0] = -1.2465; FX[5][0] = -5.8467; FX[6][0] = -9.8856; FX[7][0] = -6.5455; FX[8][0] = -1.3280; FX[9][0] = -0.0998; FX[10][0] = 0.5673; FX[11][0] = 4.3780; FX[12][0] = 9.9982; FX[13][0] = 7.9058; FX[14][0] = 2.1135; FX[15][0] = 0.0445; FX[16][0] = -0.2031; FX[17][0] = -2.6644; FX[18][0] = -8.5191; FX[19][0] = -9.3558; FX[20][0] = -3.7042; FX[21][0] = -0.4917; FX[22][0] = 0.1467; FX[23][0] = 1.5476; FX[24][0] = 6.1751; FX[25][0] = 9.9560; FX[26][0] = 6.1839; FX[27][0] = 1.0408; FX[28][0] = -0.0832; FX[29][0] = -1.0267; FX[30][0] = -5.6097; FX[31][0] = -9.6530; FX[32][0] = -7.0017; FX[33][0] = -1.6708; FX[34][0] = -0.0254; FX[35][0] = 0.4958; FX[0][1] = 9.7692; FX[1][1] = 4.7841; FX[2][1] = 0.7512; FX[3][1] = -0.0187; FX[4][1] = -1.3364; FX[5][1] = -6.0001; FX[6][1] = -9.9769; FX[7][1] = -6.2665; FX[8][1] = -1.3125; FX[9][1] = -0.1239; FX[10][1] = 0.5495; FX[11][1] = 4.5197; FX[12][1] = 10.0065; FX[13][1] = 7.9799; FX[14][1] = 1.9510; FX[15][1] = 0.0474; FX[16][1] = -0.2392; FX[17][1] = -2.8272; FX[18][1] = -8.6542; FX[19][1] = -9.1839; FX[20][1] = -3.5316; FX[21][1] = -0.4825; FX[22][1] = 0.2431; FX[23][1] = 1.3411; FX[24][1] = 5.8549; FX[25][1] = 9.7807; FX[26][1] = 6.4327; FX[27][1] = 1.3130; FX[28][1] = -0.0132; FX[29][1] = -0.8884; FX[30][1] = -5.1039; FX[31][1] = -9.8677; FX[32][1] = -7.3191; FX[33][1] = -

1.7988; FX[34][1] = -0.0307; FX[35][1] = 0.4982; FX[0][2] = 10.4460; FX[1][2] = 4.5908;
 FX[2][2] = 0.6198; FX[3][2] = -0.0529; FX[4][2] = -1.5348; FX[5][2] = -6.3648; FX[6][2]
 = -9.8983; FX[7][2] = -6.0233; FX[8][2] = -1.0017; FX[9][2] = -0.1664; FX[10][2] =
 0.5557; FX[11][2] = 4.4414; FX[12][2] = 10.0848; FX[13][2] = 8.0269; FX[14][2] =
 1.8669; FX[15][2] = 0.1109; FX[16][2] = -0.1475; FX[17][2] = -2.6881; FX[18][2] =
 -8.6186; FX[19][2] = -9.2515; FX[20][2] = -3.7271; FX[21][2] = -0.4767; FX[22][2]
 = 0.1389; FX[23][2] = 1.4849; FX[24][2] = 6.0479; FX[25][2] = 9.8146; FX[26][2] =
 6.1450; FX[27][2] = 1.2402; FX[28][2] = 0.0406; FX[29][2] = -0.6873; FX[30][2] =
 -4.8774; FX[31][2] = -9.9026; FX[32][2] = -7.5017; FX[33][2] = -1.9240; FX[34][2]
 = -0.1561; FX[35][2] = 0.3799; FX[0][3] = 10.5195; FX[1][3] = 5.0928; FX[2][3] =
 0.7401; FX[3][3] = -0.1002; FX[4][3] = -1.3206; FX[5][3] = -6.2522; FX[6][3] = -10.0288;
 FX[7][3] = -6.0361; FX[8][3] = -1.1723; FX[9][3] = -0.1206; FX[10][3] = 0.5222; FX[11][3]
 = 4.3504; FX[12][3] = 10.0915; FX[13][3] = 8.1599; FX[14][3] = 1.8739; FX[15][3]
 = 0.0503; FX[16][3] = -0.0876; FX[17][3] = -2.3910; FX[18][3] = -8.5521; FX[19][3]
 = -9.5752; FX[20][3] = -3.8999; FX[21][3] = -0.5121; FX[22][3] = 0.1466; FX[23][3]
 = 1.4441; FX[24][3] = 6.4537; FX[25][3] = 10.2133; FX[26][3] = 5.8146; FX[27][3]
 = 0.9333; FX[28][3] = 0.0204; FX[29][3] = -0.4858; FX[30][3] = -4.8187; FX[31][3]
 = -9.8864; FX[32][3] = -7.5187; FX[33][3] = -2.0924; FX[34][3] = -0.2694; FX[35][3]
 = 0.2642; FX[0][4] = 10.4588; FX[1][4] = 5.8293; FX[2][4] = 1.0247; FX[3][4] = -
 0.0113; FX[4][4] = -0.8055; FX[5][4] = -5.6403; FX[6][4] = -10.1306; FX[7][4] = -
 6.6565; FX[8][4] = -1.5953; FX[9][4] = -0.1178; FX[10][4] = 0.4177; FX[11][4] = 4.1548;
 FX[12][4] = 9.7679; FX[13][4] = 8.3656; FX[14][4] = 2.2037; FX[15][4] = 0.0807; FX[16][4]
 = -0.1240; FX[17][4] = -2.4876; FX[18][4] = -8.6345; FX[19][4] = -9.6607; FX[20][4]
 = -3.8111; FX[21][4] = -0.3790; FX[22][4] = 0.1923; FX[23][4] = 1.3777; FX[24][4]
 = 6.7002; FX[25][4] = 10.4340; FX[26][4] = 5.6433; FX[27][4] = 0.7060; FX[28][4]
 = 0.0362; FX[29][4] = -0.4477; FX[30][4] = -4.8906; FX[31][4] = -9.7504; FX[32][4]
 = -7.5512; FX[33][4] = -2.1112; FX[34][4] = -0.3108; FX[35][4] = 0.2829; FX[0][5]

= 10.4154; FX[1][5] = 5.8198; FX[2][5] = 0.9381; FX[3][5] = 0.0312; FX[4][5] = -
 0.5344; FX[5][5] = -5.3545; FX[6][5] = -10.1778; FX[7][5] = -6.8937; FX[8][5] = -
 1.8210; FX[9][5] = -0.2013; FX[10][5] = 0.4484; FX[11][5] = 3.7881; FX[12][5] = 9.4633;
 FX[13][5] = 8.4928; FX[14][5] = 2.5657; FX[15][5] = 0.2262; FX[16][5] = -0.1445;
 FX[17][5] = -2.8688; FX[18][5] = -9.0304; FX[19][5] = -9.4067; FX[20][5] = -3.3964;
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 FX[1][6] = 5.4280; FX[2][6] = 0.7330; FX[3][6] = 0.0418; FX[4][6] = -0.6276; FX[5][6]
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 FX[19][7] = -9.2376; FX[20][7] = -3.5606; FX[21][7] = -0.4576; FX[22][7] = -0.0156;
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 = -0.3670; FX[34][19] = 0.0600; FX[35][19] = 1.2186; FX[0][20] = 1.5237; FX[1][20]
 = 0.0941; FX[2][20] = -1.2989; FX[3][20] = -5.1303; FX[4][20] = -8.4197; FX[5][20] =

-6.5224; FX[6][20] = -2.6117; FX[7][20] = -0.6066; FX[8][20] = -0.1210; FX[9][20] = 1.0704; FX[10][20] = 5.0089; FX[11][20] = 9.0137; FX[12][20] = 7.0505; FX[13][20] = 2.3066; FX[14][20] = 0.3005; FX[15][20] = 0.0200; FX[16][20] = -1.9080; FX[17][20] = -7.2241; FX[18][20] = -9.4997; FX[19][20] = -5.0876; FX[20][20] = -1.0413; FX[21][20] = 0.1861; FX[22][20] = 1.0093; FX[23][20] = 5.3017; FX[24][20] = 9.3758; FX[25][20] = 6.9304; FX[26][20] = 1.7258; FX[27][20] = 0.2155; FX[28][20] = -0.3940; FX[29][20] = -3.3432; FX[30][20] = -9.0569; FX[31][20] = -8.9663; FX[32][20] = -3.0687; FX[33][20] = -0.2165; FX[34][20] = 0.0551; FX[35][20] = 1.0772; FX[0][21] = 1.3027; FX[1][21] = -0.3658; FX[2][21] = -3.1245; FX[3][21] = -7.3131; FX[4][21] = -7.9460; FX[5][21] = -4.2988; FX[6][21] = -1.2946; FX[7][21] = -0.2524; FX[8][21] = 0.3814; FX[9][21] = 2.3465; FX[10][21] = 6.7873; FX[11][21] = 8.9785; FX[12][21] = 5.1041; FX[13][21] = 1.0867; FX[14][21] = 0.0714; FX[15][21] = -0.1842; FX[16][21] = -2.8573; FX[17][21] = -7.6105; FX[18][21] = -8.7907; FX[19][21] = -4.3254; FX[20][21] = -1.0110; FX[21][21] = 0.1281; FX[22][21] = 1.3260; FX[23][21] = 5.9444; FX[24][21] = 9.5637; FX[25][21] = 6.2782; FX[26][21] = 1.4186; FX[27][21] = 0.1201; FX[28][21] = -0.4239; FX[29][21] = -4.1292; FX[30][21] = -9.6692; FX[31][21] = -8.2136; FX[32][21] = -2.4905; FX[33][21] = -0.1537; FX[34][21] = 0.0757; FX[35][21] = 1.2948; FX[0][22] = 0.6375; FX[1][22] = -1.7843; FX[2][22] = -5.4721; FX[3][22] = -7.8791; FX[4][22] = -6.2580; FX[5][22] = -2.4727; FX[6][22] = -0.5015; FX[7][22] = -0.0237; FX[8][22] = 1.3232; FX[9][22] = 4.6706; FX[10][22] = 7.9686; FX[11][22] = 7.4512; FX[12][22] = 3.0308; FX[13][22] = 0.3489; FX[14][22] = 0.0329; FX[15][22] = -0.5739; FX[16][22] = -3.8650; FX[17][22] = -8.0939; FX[18][22] = -8.0102; FX[19][22] = -3.5207; FX[20][22] = -0.7740; FX[21][22] = 0.1121; FX[22][22] = 1.8578; FX[23][22] = 6.5944; FX[24][22] = 9.4263; FX[25][22] = 5.6389; FX[26][22] = 1.1656; FX[27][22] = 0.0374; FX[28][22] = -0.4749; FX[29][22] = -5.0113; FX[30][22] = -10.1183; FX[31][22] = -7.4440; FX[32][22] = -1.9034; FX[33][22] = -0.1359; FX[34][22] = 0.1181; FX[35][22] = 1.9147; FX[0][23] = -1.1385; FX[1][23] = -4.2587; FX[2][23] = -6.7345; FX[3][23] = -6.4762; FX[4][23] = -3.6693; FX[5][23]

= -1.0575; FX[6][23] = -0.0094; FX[7][23] = 0.7815; FX[8][23] = 3.5780; FX[9][23] = 7.0849; FX[10][23] = 7.4768; FX[11][23] = 4.5213; FX[12][23] = 1.3495; FX[13][23] = 0.0594; FX[14][23] = -0.1622; FX[15][23] = -1.4019; FX[16][23] = -5.0548; FX[17][23] = -8.4397; FX[18][23] = -6.8616; FX[19][23] = -2.4241; FX[20][23] = -0.5224; FX[21][23] = 0.2020; FX[22][23] = 2.2406; FX[23][23] = 7.2235; FX[24][23] = 9.2742; FX[25][23] = 5.0217; FX[26][23] = 0.8536; FX[27][23] = 0.0790; FX[28][23] = -0.6543; FX[29][23] = -5.8341; FX[30][23] = -10.5466; FX[31][23] = -6.6366; FX[32][23] = -1.3431; FX[33][23] = -0.0635; FX[34][23] = 0.2208; FX[35][23] = 2.1243; FX[0][24] = -3.6671; FX[1][24] = -5.7917; FX[2][24] = -5.3815; FX[3][24] = -3.5718; FX[4][24] = -1.3263; FX[5][24] = -0.2840; FX[6][24] = 0.6585; FX[7][24] = 3.0001; FX[8][24] = 6.4538; FX[9][24] = 7.7593; FX[10][24] = 4.9218; FX[11][24] = 1.6891; FX[12][24] = 0.3811; FX[13][24] = 0.0185; FX[14][24] = -0.7517; FX[15][24] = -2.9590; FX[16][24] = -6.2763; FX[17][24] = -7.6911; FX[18][24] = -5.3663; FX[19][24] = -1.6524; FX[20][24] = -0.2051; FX[21][24] = 0.5309; FX[22][24] = 2.8602; FX[23][24] = 7.7616; FX[24][24] = 8.9281; FX[25][24] = 4.1802; FX[26][24] = 0.5631; FX[27][24] = 0.1050; FX[28][24] = -0.9101; FX[29][24] = -6.3146; FX[30][24] = -10.7937; FX[31][24] = -6.1028; FX[32][24] = -0.9610; FX[33][24] = 0.0059; FX[34][24] = 0.2731; FX[35][24] = 2.2844; FX[0][25] = -5.4960; FX[1][25] = -4.5374; FX[2][25] = -2.2625; FX[3][25] = -1.0133; FX[4][25] = -0.2730; FX[5][25] = 0.4292; FX[6][25] = 2.6132; FX[7][25] = 5.8314; FX[8][25] = 7.5307; FX[9][25] = 5.7524; FX[10][25] = 2.0408; FX[11][25] = 0.3405; FX[12][25] = 0.1006; FX[13][25] = -0.3261; FX[14][25] = -2.2836; FX[15][25] = -5.1189; FX[16][25] = -6.6192; FX[17][25] = -5.9780; FX[18][25] = -3.5042; FX[19][25] = -1.0044; FX[20][25] = 0.1611; FX[21][25] = 1.1544; FX[22][25] = 3.7210; FX[23][25] = 7.7916; FX[24][25] = 8.1679; FX[25][25] = 3.4328; FX[26][25] = 0.3280; FX[27][25] = 0.0602; FX[28][25] = -0.9680; FX[29][25] = -6.3525; FX[30][25] = -10.7104; FX[31][25] = -6.0608; FX[32][25] = -0.9543; FX[33][25] = -0.0245; FX[34][25] = 0.2466; FX[35][25] = 2.4133; FX[0][26] = -4.2683; FX[1][26] = -1.8562; FX[2][26] = -0.4053; FX[3][26] = 0.0483; FX[4][26] = 0.6423; FX[5][26]

= 2.4452; FX[6][26] = 5.7322; FX[7][26] = 7.1331; FX[8][26] = 5.4097; FX[9][26] = 2.6169; FX[10][26] = 0.4430; FX[11][26] = -0.0135; FX[12][26] = -0.4773; FX[13][26] = -2.1037; FX[14][26] = -4.5611; FX[15][26] = -6.4424; FX[16][26] = -5.2609; FX[17][26] = -3.3211; FX[18][26] = -1.7567; FX[19][26] = -0.3011; FX[20][26] = 0.5171; FX[21][26] = 1.6010; FX[22][26] = 3.9114; FX[23][26] = 7.3593; FX[24][26] = 7.5071; FX[25][26] = 3.1260; FX[26][26] = 0.2507; FX[27][26] = -0.0075; FX[28][26] = -0.9108; FX[29][26] = -6.0902; FX[30][26] = -10.3904; FX[31][26] = -6.4487; FX[32][26] = -1.1658; FX[33][26] = -0.0438; FX[34][26] = 0.1972; FX[35][26] = 2.3218; FX[0][27] = -2.1803; FX[1][27] = -0.2797; FX[2][27] = 0.2388; FX[3][27] = 0.7685; FX[4][27] = 2.5850; FX[5][27] = 5.7401; FX[6][27] = 7.3153; FX[7][27] = 5.3478; FX[8][27] = 2.1462; FX[9][27] = 0.5795; FX[10][27] = -0.0771; FX[11][27] = -0.9292; FX[12][27] = -2.8222; FX[13][27] = -4.7746; FX[14][27] = -5.6488; FX[15][27] = -4.9213; FX[16][27] = -2.7282; FX[17][27] = -1.1897; FX[18][27] = -0.2984; FX[19][27] = 0.3646; FX[20][27] = 0.7803; FX[21][27] = 1.5125; FX[22][27] = 3.3700; FX[23][27] = 6.8109; FX[24][27] = 7.0841; FX[25][27] = 3.0930; FX[26][27] = 0.3855; FX[27][27] = 0.0451; FX[28][27] = -0.9254; FX[29][27] = -5.7871; FX[30][27] = -10.2676; FX[31][27] = -6.8167; FX[32][27] = -1.2562; FX[33][27] = -0.0300; FX[34][27] = 0.2422; FX[35][27] = 2.0080; FX[0][28] = -0.6184; FX[1][28] = 0.3327; FX[2][28] = 1.0695; FX[3][28] = 2.6528; FX[4][28] = 5.5936; FX[5][28] = 7.0484; FX[6][28] = 5.6131; FX[7][28] = 2.2499; FX[8][28] = 0.3799; FX[9][28] = -0.0538; FX[10][28] = -1.1718; FX[11][28] = -3.5745; FX[12][28] = -5.9034; FX[13][28] = -5.8291; FX[14][28] = -3.9340; FX[15][28] = -2.1278; FX[16][28] = -0.6413; FX[17][28] = 0.2259; FX[18][28] = 0.7940; FX[19][28] = 0.9751; FX[20][28] = 0.8115; FX[21][28] = 1.0604; FX[22][28] = 2.4820; FX[23][28] = 5.7793; FX[24][28] = 6.6074; FX[25][28] = 3.5762; FX[26][28] = 0.7886; FX[27][28] = 0.1758; FX[28][28] = -0.8622; FX[29][28] = -5.4882; FX[30][28] = -10.1551; FX[31][28] = -6.9544; FX[32][28] = -1.5410; FX[33][28] = -0.0726; FX[34][28] = 0.1899; FX[35][28] = 1.5107; FX[0][29] = 0.1163; FX[1][29] = 0.9277; FX[2][29] = 3.1115; FX[3][29] = 5.7755; FX[4][29] = 6.8268; FX[5][29] =

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 = 1.5359; FX[18][29] = 1.5321; FX[19][29] = 0.8210; FX[20][29] = 0.2062; FX[21][29] =
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 = 2.3642; FX[2][30] = 5.9826; FX[3][30] = 7.7081; FX[4][30] = 5.6392; FX[5][30] =
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 = 2.9513; FX[18][30] = 1.6612; FX[19][30] = -0.3135; FX[20][30] = -1.2430; FX[21][30]
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 = -0.5467; FX[34][30] = -0.0188; FX[35][30] = 0.6714; FX[0][31] = 1.5554; FX[1][31]
 = 4.6291; FX[2][31] = 7.9511; FX[3][31] = 6.9821; FX[4][31] = 3.2972; FX[5][31] =
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-2.3238; FX[6][38] = -7.5433; FX[7][38] = -9.5122; FX[8][38] = -4.5289; FX[9][38] =
 -0.7520; FX[10][38] = -0.0347; FX[11][38] = 0.6915; FX[12][38] = 5.7902; FX[13][38] =
 10.8015; FX[14][38] = 6.7868; FX[15][38] = 1.0459; FX[16][38] = -0.0513; FX[17][38] =
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 = -4.3418; FX[22][38] = -0.2675; FX[23][38] = 0.3981; FX[24][38] = 2.9554; FX[25][38]
 = 8.2636; FX[26][38] = 9.0028; FX[27][38] = 3.8889; FX[28][38] = 0.5342; FX[29][38] =
 -0.2350; FX[30][38] = -2.5574; FX[31][38] = -8.5598; FX[32][38] = -9.5083; FX[33][38]
 = -3.9211; FX[34][38] = -0.2657; FX[35][38] = 0.0802; FX[0][39] = 9.9083; FX[1][39]
 = 6.9183; FX[2][39] = 1.6686; FX[3][39] = -0.0016; FX[4][39] = -0.5360; FX[5][39] =
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 10.8891; FX[14][39] = 5.3871; FX[15][39] = 0.3955; FX[16][39] = -0.0310; FX[17][39] =
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 = -2.9120; FX[22][39] = -0.0844; FX[23][39] = 0.3479; FX[24][39] = 2.9586; FX[25][39]
 = 8.5948; FX[26][39] = 9.2612; FX[27][39] = 3.5275; FX[28][39] = 0.3958; FX[29][39] =
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 = -3.6227; FX[34][39] = -0.2620; FX[35][39] = 0.1380; FX[0][40] = 9.9859; FX[1][40]
 = 6.4352; FX[2][40] = 1.3505; FX[3][40] = -0.0150; FX[4][40] = -0.5635; FX[5][40] =
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 = -1.7676; FX[22][40] = 0.0463; FX[23][40] = 0.4112; FX[24][40] = 3.3063; FX[25][40]
 = 8.9285; FX[26][40] = 8.9501; FX[27][40] = 3.1226; FX[28][40] = 0.2989; FX[29][40] =
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 = 6.1826; FX[2][41] = 1.0802; FX[3][41] = -0.0496; FX[4][41] = -0.5477; FX[5][41] =

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 = -1.1747; FX[22][41] = 0.0201; FX[23][41] = 0.4099; FX[24][41] = 3.9281; FX[25][41]
 = 9.5323; FX[26][41] = 8.4206; FX[27][41] = 2.4988; FX[28][41] = 0.1602; FX[29][41] =
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 = 6.0646; FX[2][42] = 1.0681; FX[3][42] = -0.0346; FX[4][42] = -0.7151; FX[5][42] =
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 = -0.9150; FX[22][42] = 0.0104; FX[23][42] = 0.5297; FX[24][42] = 4.6936; FX[25][42]
 = 9.7662; FX[26][42] = 7.6721; FX[27][42] = 2.1749; FX[28][42] = 0.1052; FX[29][42] =
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 = 6.0273; FX[2][43] = 1.1278; FX[3][43] = -0.0204; FX[4][43] = -0.8956; FX[5][43] =
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 = -0.6356; FX[22][43] = 0.0648; FX[23][43] = 0.9439; FX[24][43] = 5.2212; FX[25][43]
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 = 6.4808; FX[2][44] = 1.0179; FX[3][44] = -0.0295; FX[4][44] = -1.0050; FX[5][44] =

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 = 9.6351; FX[26][44] = 6.3461; FX[27][44] = 1.8280; FX[28][44] = 0.2079; FX[29][44] =
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 = 7.0116; FX[2][45] = 1.0569; FX[3][45] = -0.0987; FX[4][45] = -0.8745; FX[5][45] =
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 = 9.4725; FX[26][45] = 6.2287; FX[27][45] = 1.5943; FX[28][45] = 0.1514; FX[29][45] =
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 = 9.4273; FX[26][46] = 6.4719; FX[27][46] = 1.7015; FX[28][46] = 0.0500; FX[29][46] =
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 = 6.8108; FX[2][47] = 1.1110; FX[3][47] = 0.0345; FX[4][47] = -0.3396; FX[5][47] =

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 = 9.5701; FX[26][47] = 6.8477; FX[27][47] = 1.5347; FX[28][47] = 0.0125; FX[29][47] =
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 = 6.9277; FX[2][48] = 1.2860; FX[3][48] = 0.0696; FX[4][48] = -0.3491; FX[5][48] =
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 = 0.2894; V[20][3] = 0.8455; V[21][3] = 0.9865; V[22][3] = 0.9977; V[23][3] = 0.9465;
 V[24][3] = 0.6666; V[25][3] = -0.0618; V[26][3] = -0.7429; V[27][3] = -0.9838; V[28][3]
 = -1.0029; V[29][3] = -0.9959; V[30][3] = -0.8242; V[31][3] = -0.1960; V[32][3] = 0.5553;

$V[33][3] = 0.9245$; $V[34][3] = 0.9962$; $V[35][3] = 0.9943$; $V[0][4] = -0.0408$; $V[1][4] = -0.7328$; $V[2][4] = -0.9787$; $V[3][4] = -1.0013$; $V[4][4] = -0.9853$; $V[5][4] = -0.7605$; $V[6][4] = -0.0849$; $V[7][4] = 0.6399$; $V[8][4] = 0.9455$; $V[9][4] = 0.9931$; $V[10][4] = 0.9883$; $V[11][4] = 0.8428$; $V[12][4] = 0.2648$; $V[13][4] = -0.5317$; $V[14][4] = -0.9432$; $V[15][4] = -1.0029$; $V[16][4] = -1.0032$; $V[17][4] = -0.9306$; $V[18][4] = -0.4902$; $V[19][4] = 0.3088$; $V[20][4] = 0.8676$; $V[21][4] = 0.9974$; $V[22][4] = 0.9991$; $V[23][4] = 0.9540$; $V[24][4] = 0.6624$; $V[25][4] = -0.0801$; $V[26][4] = -0.7730$; $V[27][4] = -0.9873$; $V[28][4] = -1.0013$; $V[29][4] = -0.9977$; $V[30][4] = -0.8216$; $V[31][4] = -0.2019$; $V[32][4] = 0.5522$; $V[33][4] = 0.9178$; $V[34][4] = 0.9958$; $V[35][4] = 0.9926$; $V[0][5] = -0.0498$; $V[1][5] = -0.7423$; $V[2][5] = -0.9816$; $V[3][5] = -1.0029$; $V[4][5] = -0.9985$; $V[5][5] = -0.7986$; $V[6][5] = -0.1356$; $V[7][5] = 0.6052$; $V[8][5] = 0.9291$; $V[9][5] = 0.9895$; $V[10][5] = 0.9855$; $V[11][5] = 0.8519$; $V[12][5] = 0.3021$; $V[13][5] = -0.4798$; $V[14][5] = -0.9196$; $V[15][5] = -0.9994$; $V[16][5] = -1.0046$; $V[17][5] = -0.9170$; $V[18][5] = -0.4408$; $V[19][5] = 0.3682$; $V[20][5] = 0.8910$; $V[21][5] = 0.9996$; $V[22][5] = 0.9985$; $V[23][5] = 0.9631$; $V[24][5] = 0.6662$; $V[25][5] = -0.0737$; $V[26][5] = -0.7636$; $V[27][5] = -0.9784$; $V[28][5] = -1.0003$; $V[29][5] = -0.9954$; $V[30][5] = -0.8179$; $V[31][5] = -0.1890$; $V[32][5] = 0.5676$; $V[33][5] = 0.9292$; $V[34][5] = 1.0010$; $V[35][5] = 0.9938$; $V[0][6] = -0.1003$; $V[1][6] = -0.7743$; $V[2][6] = -0.9850$; $V[3][6] = -1.0029$; $V[4][6] = -0.9938$; $V[5][6] = -0.7894$; $V[6][6] = -0.1351$; $V[7][6] = 0.6008$; $V[8][6] = 0.9199$; $V[9][6] = 0.9871$; $V[10][6] = 0.9828$; $V[11][6] = 0.8368$; $V[12][6] = 0.2921$; $V[13][6] = -0.4811$; $V[14][6] = -0.9243$; $V[15][6] = -1.0009$; $V[16][6] = -1.0046$; $V[17][6] = -0.9145$; $V[18][6] = -0.4214$; $V[19][6] = 0.3848$; $V[20][6] = 0.8869$; $V[21][6] = 0.9926$; $V[22][6] = 0.9988$; $V[23][6] = 0.9673$; $V[24][6] = 0.6747$; $V[25][6] = -0.0569$; $V[26][6] = -0.7411$; $V[27][6] = -0.9757$; $V[28][6] = -1.0022$; $V[29][6] = -0.9980$; $V[30][6] = -0.8259$; $V[31][6] = -0.1905$; $V[32][6] = 0.5926$; $V[33][6] = 0.9521$; $V[34][6] = 1.0017$; $V[35][6] = 0.9933$; $V[0][7] = -0.1167$; $V[1][7] = -0.7788$; $V[2][7] = -0.9841$; $V[3][7] = -1.0013$; $V[4][7] = -0.9834$; $V[5][7] = -0.7682$; $V[6][7] = -0.1277$; $V[7][7] = 0.5839$; $V[8][7] = 0.9171$; $V[9][7] = 0.9866$; $V[10][7] =$

0.9826; $V[11][7] = 0.8418$; $V[12][7] = 0.2927$; $V[13][7] = -0.5075$; $V[14][7] = -0.9376$;
 $V[15][7] = -1.0042$; $V[16][7] = -1.0048$; $V[17][7] = -0.9317$; $V[18][7] = -0.4563$; $V[19][7]$
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 $= -1.0029$; $V[29][7] = -0.9971$; $V[30][7] = -0.8212$; $V[31][7] = -0.1661$; $V[32][7] = 0.6251$;
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 -0.8065 ; $V[2][9] = -0.9796$; $V[3][9] = -0.9972$; $V[4][9] = -0.9690$; $V[5][9] = -0.7449$;
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 -0.8437 ; $V[2][10] = -0.9943$; $V[3][10] = -1.0026$; $V[4][10] = -0.9844$; $V[5][10] = -0.7546$;
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 -1.0025 ; $V[29][11] = -0.9857$; $V[30][11] = -0.7048$; $V[31][11] = 0.0602$; $V[32][11] =$
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 $= -0.6810$; $V[6][12] = -0.0124$; $V[7][12] = 0.6616$; $V[8][12] = 0.9501$; $V[9][12] = 0.9967$;
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 $= -0.9267$; $V[15][12] = -0.9982$; $V[16][12] = -1.0024$; $V[17][12] = -0.8696$; $V[18][12]$
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 $= -1.0025$; $V[4][13] = -0.9590$; $V[5][13] = -0.6657$; $V[6][13] = 0.0155$; $V[7][13] = 0.6822$;
 $V[8][13] = 0.9528$; $V[9][13] = 0.9988$; $V[10][13] = 0.9973$; $V[11][13] = 0.8525$; $V[12][13]$
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 $= -0.2952$; $V[14][45] = -0.8397$; $V[15][45] = -0.9947$; $V[16][45] = -1.0050$; $V[17][45]$

= -0.9592; V[18][45] = -0.6277; V[19][45] = 0.1398; V[20][45] = 0.7944; V[21][45] = 0.9918; V[22][45] = 0.9977; V[23][45] = 0.9543; V[24][45] = 0.6779; V[25][45] = 0.0156; V[26][45] = -0.6586; V[27][45] = -0.9501; V[28][45] = -1.0001; V[29][45] = -0.9986; V[30][45] = -0.8583; V[31][45] = -0.3179; V[32][45] = 0.4343; V[33][45] = 0.8963; V[34][45] = 0.9998; V[35][45] = 0.9991; V[0][46] = 0.0712; V[1][46] = -0.6903; V[2][46] = -0.9839; V[3][46] = -1.0025; V[4][46] = -0.9902; V[5][46] = -0.8359; V[6][46] = -0.2473; V[7][46] = 0.5185; V[8][46] = 0.9261; V[9][46] = 0.9949; V[10][46] = 0.9920; V[11][46] = 0.9194; V[12][46] = 0.5206; V[13][46] = -0.2432; V[14][46] = -0.8273; V[15][46] = -0.9950; V[16][46] = -1.0060; V[17][46] = -0.9558; V[18][46] = -0.6234; V[19][46] = 0.1212; V[20][46] = 0.7712; V[21][46] = 0.9872; V[22][46] = 0.9974; V[23][46] = 0.9416; V[24][46] = 0.6759; V[25][46] = 0.0354; V[26][46] = -0.6456; V[27][46] = -0.9591; V[28][46] = -1.0035; V[29][46] = -0.9967; V[30][46] = -0.8413; V[31][46] = -0.2952; V[32][46] = 0.4422; V[33][46] = 0.8901; V[34][46] = 1.0001; V[35][46] = 0.9977; V[0][47] = 0.0610; V[1][47] = -0.6888; V[2][47] = -0.9791; V[3][47] = -1.0024; V[4][47] = -0.9997; V[5][47] = -0.8635; V[6][47] = -0.3020; V[7][47] = 0.4770; V[8][47] = 0.9078; V[9][47] = 0.9887; V[10][47] = 0.9949; V[11][47] = 0.9425; V[12][47] = 0.5838; V[13][47] = -0.1833; V[14][47] = -0.8123; V[15][47] = -0.9960; V[16][47] = -1.0052; V[17][47] = -0.9688; V[18][47] = -0.6479; V[19][47] = 0.0986; V[20][47] = 0.7674; V[21][47] = 0.9894; V[22][47] = 0.9990; V[23][47] = 0.9528; V[24][47] = 0.6980; V[25][47] = 0.0587; V[26][47] = -0.6468; V[27][47] = -0.9691; V[28][47] = -1.0028; V[29][47] = -0.9963; V[30][47] = -0.8310; V[31][47] = -0.2609; V[32][47] = 0.4838; V[33][47] = 0.9101; V[34][47] = 1.0029; V[35][47] = 0.9988; V[0][48] = 0.0911; V[1][48] = -0.6669; V[2][48] = -0.9702; V[3][48] = -1.0022; V[4][48] = -0.9990; V[5][48] = -0.8680; V[6][48] = -0.3506; V[7][48] = 0.4090; V[8][48] = 0.8769; V[9][48] = 0.9871; V[10][48] = 0.9971; V[11][48] = 0.9575; V[12][48] = 0.6291; V[13][48] = -0.1311; V[14][48] = -0.7826; V[15][48] = -0.9944; V[16][48] = -1.0034; V[17][48] = -0.9675; V[18][48] = -0.6642; V[19][48] = 0.0802; V[20][48] = 0.7620; V[21][48] = 0.9892; V[22][48] = 0.9947; V[23][48] = 0.9947;

= 0.9583; V[24][48] = 0.7259; V[25][48] = 0.0663; V[26][48] = -0.6626; V[27][48] = -0.9756; V[28][48] = -1.0017; V[29][48] = -0.9935; V[30][48] = -0.8309; V[31][48] = -0.2674; V[32][48] = 0.5000; V[33][48] = 0.9233; V[34][48] = 1.0025; V[35][48] = 0.9997; V[0][49] = 0.1475; V[1][49] = -0.6299; V[2][49] = -0.9710; V[3][49] = -1.0024; V[4][49] = -1.0022; V[5][49] = -0.8958; V[6][49] = -0.4179; V[7][49] = 0.3269; V[8][49] = 0.8350; V[9][49] = 0.9812; V[10][49] = 0.9997; V[11][49] = 0.9609; V[12][49] = 0.6196; V[13][49] = -0.1454; V[14][49] = -0.8012; V[15][49] = -0.9961; V[16][49] = -1.0034; V[17][49] = -0.9554; V[18][49] = -0.6382; V[19][49] = 0.0822; V[20][49] = 0.7405; V[21][49] = 0.9755; V[22][49] = 0.9898; V[23][49] = 0.9541; V[24][49] = 0.7367; V[25][49] = 0.0939; V[26][49] = -0.6428; V[27][49] = -0.9693; V[28][49] = -1.0036; V[29][49] = -0.9901; V[30][49] = -0.8476; V[31][49] = -0.2788; V[32][49] = 0.5053; V[33][49] = 0.9306; V[34][49] = 1.0000; V[35][49] = 0.9990; V[0][50] = 0.1715; V[1][50] = -0.6115; V[2][50] = -0.9612; V[3][50] = -0.9992; V[4][50] = -1.0049; V[5][50] = -0.9171; V[6][50] = -0.4524; V[7][50] = 0.2973; V[8][50] = 0.8069; V[9][50] = 0.9716; V[10][50] = 1.0001; V[11][50] = 0.9642; V[12][50] = 0.6139; V[13][50] = -0.1581; V[14][50] = -0.8159; V[15][50] = -0.9992; V[16][50] = -1.0032; V[17][50] = -0.9540; V[18][50] = -0.6321; V[19][50] = 0.0707; V[20][50] = 0.7147; V[21][50] = 0.9596; V[22][50] = 0.9951; V[23][50] = 0.9681; V[24][50] = 0.7615; V[25][50] = 0.1184; V[26][50] = -0.6302; V[27][50] = -0.9688; V[28][50] = -1.0041; V[29][50] = -0.9958; V[30][50] = -0.8466; V[31][50] = -0.2588; V[32][50] = 0.5389; V[33][50] = 0.9432; V[34][50] = 0.9990; V[35][50] = 0.9999; V[0][51] = 0.1700; V[1][51] = -0.6014; V[2][51] = -0.9504; V[3][51] = -0.9973; V[4][51] = -1.0033; V[5][51] = -0.9242; V[6][51] = -0.4431; V[7][51] = 0.3129; V[8][51] = 0.8040; V[9][51] = 0.9609; V[10][51] = 0.9985; V[11][51] = 0.9645; V[12][51] = 0.6621; V[13][51] = -0.0938; V[14][51] = -0.7811; V[15][51] = -0.9967; V[16][51] = -1.0034; V[17][51] = -0.9675; V[18][51] = -0.6789; V[19][51] = -0.0023; V[20][51] = 0.6574; V[21][51] = 0.9473; V[22][51] = 0.9960; V[23][51] = 0.9780; V[24][51] = 0.7691; V[25][51] = 0.0995; V[26][51] = -0.6522; V[27][51] = -0.9748; V[28][51] = -1.0035; V[29][51] =

-0.9998; V[30][51] = -0.8471; V[31][51] = -0.2449; V[32][51] = 0.5526; V[33][51] = 0.9473; V[34][51] = 1.0010; V[35][51] = 0.9996;

B.0.5 Defect Order = 5

$if(m_{index} == 1 \&\& m_{pos.x} > (-3 * pitch) \&\& m_{pos.x} < (3 * pitch) \&\& m_{pos.y} > (-2.0 * pitch) \&\& m_{pos.y} < (2.0 * pitch))$

FX[0][0] = 0.0349; FX[1][0] = 1.0692; FX[2][0] = 5.7961; FX[3][0] = 10.1856; FX[4][0] = 6.7587; FX[5][0] = 1.1744; FX[6][0] = -0.0319; FX[7][0] = -0.6485; FX[8][0] = -4.5864; FX[9][0] = -9.5183; FX[10][0] = -7.8746; FX[11][0] = -2.2496; FX[12][0] = -0.0935; FX[13][0] = 0.2562; FX[14][0] = 2.8360; FX[15][0] = 9.0269; FX[16][0] = 9.3359; FX[17][0] = 3.3747; FX[18][0] = 0.2119; FX[19][0] = -0.0726; FX[20][0] = -1.4954; FX[21][0] = -7.1956; FX[22][0] = -9.9064; FX[23][0] = -5.3771; FX[24][0] = -0.8763; FX[25][0] = -0.1023; FX[26][0] = 1.1486; FX[27][0] = 5.3173; FX[28][0] = 9.7711; FX[29][0] = 7.1082; FX[30][0] = 1.6538; FX[31][0] = 0.0019; FX[32][0] = -0.6714; FX[33][0] = -4.5066; FX[34][0] = -9.4007; FX[35][0] = -7.7689; FX[36][0] = -2.5202; FX[37][0] = -0.1648; FX[38][0] = 0.2274; FX[0][1] = -0.0245; FX[1][1] = 0.9280; FX[2][1] = 5.8105; FX[3][1] = 10.2135; FX[4][1] = 6.6985; FX[5][1] = 1.3973; FX[6][1] = -0.0553; FX[7][1] = -0.7062; FX[8][1] = -4.4285; FX[9][1] = -9.5276; FX[10][1] = -7.9004; FX[11][1] = -2.2485; FX[12][1] = -0.1708; FX[13][1] = 0.2552; FX[14][1] = 2.9120; FX[15][1] = 8.9526; FX[16][1] = 9.2817; FX[17][1] = 3.3330; FX[18][1] = 0.3042; FX[19][1] = -0.1400; FX[20][1] = -1.4897; FX[21][1] = -7.2051; FX[22][1] = -9.8930; FX[23][1] = -5.3099; FX[24][1] = -0.9047; FX[25][1] = -0.0072; FX[26][1] = 0.9819; FX[27][1] = 4.9040; FX[28][1] = 9.7408; FX[29][1] = 7.5089; FX[30][1] = 1.8567; FX[31][1] = -0.0428; FX[32][1] = -0.5938; FX[33][1] = -4.2502; FX[34][1] = -9.1894; FX[35][1] = -8.0753; FX[36][1] = -2.7632; FX[37][1] = -0.1561; FX[38][1] = 0.2353; FX[0][2] = -0.0454; FX[1][2] = 0.8731; FX[2][2] = 6.1142; FX[3][2] = 10.1500; FX[4][2] = 6.4946; FX[5][2] = 1.3522; FX[6][2] = 0.0650; FX[7][2] = -0.7443; FX[8][2] = -4.4663; FX[9][2]

= -9.5209; FX[10][2] = -7.8795; FX[11][2] = -2.2114; FX[12][2] = -0.2391; FX[13][2] = 0.2478; FX[14][2] = 3.1082; FX[15][2] = 9.0115; FX[16][2] = 9.3121; FX[17][2] = 3.1570; FX[18][2] = 0.2376; FX[19][2] = -0.1551; FX[20][2] = -1.7089; FX[21][2] = -7.2750; FX[22][2] = -9.9569; FX[23][2] = -4.9402; FX[24][2] = -0.9162; FX[25][2] = 0.1455; FX[26][2] = 0.8624; FX[27][2] = 4.4267; FX[28][2] = 9.4130; FX[29][2] = 7.8611; FX[30][2] = 2.2369; FX[31][2] = 0.0244; FX[32][2] = -0.4444; FX[33][2] = -3.7920; FX[34][2] = -9.1972; FX[35][2] = -8.5483; FX[36][2] = -2.8878; FX[37][2] = -0.1593; FX[38][2] = 0.2706; FX[0][3] = -0.0581; FX[1][3] = 0.8419; FX[2][3] = 6.2136; FX[3][3] = 10.4952; FX[4][3] = 6.2679; FX[5][3] = 1.1695; FX[6][3] = 0.0606; FX[7][3] = -0.8382; FX[8][3] = -4.8751; FX[9][3] = -9.6711; FX[10][3] = -7.5235; FX[11][3] = -1.9041; FX[12][3] = -0.2194; FX[13][3] = 0.2299; FX[14][3] = 3.0182; FX[15][3] = 9.0440; FX[16][3] = 9.4549; FX[17][3] = 3.0809; FX[18][3] = 0.2331; FX[19][3] = -0.0480; FX[20][3] = -1.6730; FX[21][3] = -7.2422; FX[22][3] = -9.9321; FX[23][3] = -5.0795; FX[24][3] = -0.8968; FX[25][3] = 0.0476; FX[26][3] = 0.9132; FX[27][3] = 4.5129; FX[28][3] = 9.4755; FX[29][3] = 7.6347; FX[30][3] = 2.1799; FX[31][3] = 0.1330; FX[32][3] = -0.3619; FX[33][3] = -3.4845; FX[34][3] = -9.1754; FX[35][3] = -8.7467; FX[36][3] = -2.9984; FX[37][3] = -0.2832; FX[38][3] = 0.1973; FX[0][4] = -0.0660; FX[1][4] = 0.7337; FX[2][4] = 5.8840; FX[3][4] = 10.5232; FX[4][4] = 6.6149; FX[5][4] = 1.3045; FX[6][4] = -0.0252; FX[7][4] = -0.8294; FX[8][4] = -4.8124; FX[9][4] = -9.8209; FX[10][4] = -7.4265; FX[11][4] = -1.9033; FX[12][4] = -0.1971; FX[13][4] = 0.2468; FX[14][4] = 2.9449; FX[15][4] = 9.1053; FX[16][4] = 9.4787; FX[17][4] = 3.0885; FX[18][4] = 0.1815; FX[19][4] = -0.0091; FX[20][4] = -1.3762; FX[21][4] = -7.1337; FX[22][4] = -10.1207; FX[23][4] = -5.3622; FX[24][4] = -0.9337; FX[25][4] = -0.0230; FX[26][4] = 0.9724; FX[27][4] = 4.7311; FX[28][4] = 9.9757; FX[29][4] = 7.3672; FX[30][4] = 1.8556; FX[31][4] = 0.0655; FX[32][4] = -0.2100; FX[33][4] = -3.2583; FX[34][4] = -9.1999; FX[35][4] = -8.7746; FX[36][4] = -3.1642; FX[37][4] = -0.4472; FX[38][4] = 0.1188; FX[0][5] = -0.0607; FX[1][5] = 0.5107; FX[2][5] = 5.0506;

FX[3][5] = 10.4043; FX[4][5] = 7.3130; FX[5][5] = 1.7396; FX[6][5] = 0.0353; FX[7][5] = -0.4499; FX[8][5] = -4.2084; FX[9][5] = -9.6750; FX[10][5] = -7.9780; FX[11][5] = -2.4057; FX[12][5] = -0.2271; FX[13][5] = 0.1659; FX[14][5] = 2.8350; FX[15][5] = 8.7886; FX[16][5] = 9.6067; FX[17][5] = 3.3870; FX[18][5] = 0.2067; FX[19][5] = -0.0189; FX[20][5] = -1.4246; FX[21][5] = -7.0465; FX[22][5] = -10.2601; FX[23][5] = -5.4837; FX[24][5] = -0.8313; FX[25][5] = 0.0803; FX[26][5] = 0.8450; FX[27][5] = 4.9739; FX[28][5] = 10.3025; FX[29][5] = 7.3501; FX[30][5] = 1.4619; FX[31][5] = 0.0381; FX[32][5] = -0.1220; FX[33][5] = -3.3060; FX[34][5] = -9.0624; FX[35][5] = -8.8486; FX[36][5] = -3.1514; FX[37][5] = -0.5484; FX[38][5] = 0.1297; FX[0][6] = -0.0599; FX[1][6] = 0.3324; FX[2][6] = 5.0077; FX[3][6] = 10.3474; FX[4][6] = 7.5633; FX[5][6] = 1.7661; FX[6][6] = 0.0706; FX[7][6] = -0.1562; FX[8][6] = -3.7764; FX[9][6] = -9.6346; FX[10][6] = -8.3143; FX[11][6] = -2.7265; FX[12][6] = -0.3430; FX[13][6] = 0.1763; FX[14][6] = 2.4903; FX[15][6] = 8.4846; FX[16][6] = 9.5491; FX[17][6] = 3.8385; FX[18][6] = 0.4138; FX[19][6] = -0.0034; FX[20][6] = -1.6507; FX[21][6] = -7.5250; FX[22][6] = -10.2233; FX[23][6] = -5.0493; FX[24][6] = -0.6363; FX[25][6] = 0.1039; FX[26][6] = 0.7192; FX[27][6] = 5.1275; FX[28][6] = 10.2674; FX[29][6] = 7.3258; FX[30][6] = 1.4258; FX[31][6] = 0.1040; FX[32][6] = -0.1754; FX[33][6] = -3.3529; FX[34][6] = -9.1643; FX[35][6] = -8.7401; FX[36][6] = -3.1509; FX[37][6] = -0.4797; FX[38][6] = 0.1680; FX[0][7] = -0.0488; FX[1][7] = 0.4714; FX[2][7] = 5.3343; FX[3][7] = 10.5567; FX[4][7] = 7.2075; FX[5][7] = 1.4475; FX[6][7] = 0.0771; FX[7][7] = -0.2077; FX[8][7] = -3.7645; FX[9][7] = -9.4608; FX[10][7] = -8.3661; FX[11][7] = -2.6981; FX[12][7] = -0.4456; FX[13][7] = 0.2250; FX[14][7] = 2.6201; FX[15][7] = 8.2351; FX[16][7] = 9.4564; FX[17][7] = 3.9823; FX[18][7] = 0.4135; FX[19][7] = 0.0199; FX[20][7] = -1.8900; FX[21][7] = -7.7614; FX[22][7] = -10.1308; FX[23][7] = -4.6867; FX[24][7] = -0.5545; FX[25][7] = -0.0041; FX[26][7] = 0.6700; FX[27][7] = 5.0991; FX[28][7] = 10.0795; FX[29][7] = 7.3329; FX[30][7] = 1.7001; FX[31][7] = 0.1283; FX[32][7] = -0.1484; FX[33][7] = -3.2795; FX[34][7] = -9.3402; FX[35][7] =

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 = -2.8150; FX[12][9] = -0.4883; FX[13][9] = 0.1464; FX[14][9] = 2.4224; FX[15][9]
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 10.0009; FX[17][10] = 3.6680; FX[18][10] = 0.4144; FX[19][10] = 0.0307; FX[20][10] =
 -2.2380; FX[21][10] = -8.0671; FX[22][10] = -9.7248; FX[23][10] = -4.1808; FX[24][10]
 = -0.6256; FX[25][10] = 0.2409; FX[26][10] = 1.2101; FX[27][10] = 5.6821; FX[28][10]

= 9.9470; FX[29][10] = 6.2962; FX[30][10] = 1.3708; FX[31][10] = 0.1348; FX[32][10] =
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 = -2.0030; FX[37][10] = -0.0577; FX[38][10] = 0.2020; FX[0][11] = -0.0719; FX[1][11]
 = 0.7774; FX[2][11] = 6.1584; FX[3][11] = 10.9015; FX[4][11] = 6.2900; FX[5][11] =
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 = 0.2405; FX[26][11] = 1.5638; FX[27][11] = 6.4654; FX[28][11] = 9.8305; FX[29][11] =
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 = 1.7123; FX[27][12] = 6.7032; FX[28][12] = 9.9501; FX[29][12] = 5.6039; FX[30][12] =
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= -0.5827; FX[38][43] = 0.0406; FX[0][44] = -0.0288; FX[1][44] = 0.5991; FX[2][44] = 4.9019; FX[3][44] = 9.9071; FX[4][44] = 7.6236; FX[5][44] = 1.9158; FX[6][44] = 0.1056; FX[7][44] = -0.5326; FX[8][44] = -3.3745; FX[9][44] = -8.9116; FX[10][44] = -8.5607; FX[11][44] = -3.2008; FX[12][44] = -0.4083; FX[13][44] = 0.1541; FX[14][44] = 1.7792; FX[15][44] = 6.5990; FX[16][44] = 9.8305; FX[17][44] = 5.6640; FX[18][44] = 1.0078; FX[19][44] = -0.0163; FX[20][44] = -0.6547; FX[21][44] = -5.4658; FX[22][44] = -10.4526; FX[23][44] = -7.0446; FX[24][44] = -1.4368; FX[25][44] = 0.0420; FX[26][44] = 0.3850; FX[27][44] = 3.6712; FX[28][44] = 9.1395; FX[29][44] = 8.3453; FX[30][44] = 3.1578; FX[31][44] = 0.2878; FX[32][44] = -0.1326; FX[33][44] = -2.4325; FX[34][44] = -7.7941; FX[35][44] = -9.6800; FX[36][44] = -4.4534; FX[37][44] = -0.5230; FX[38][44] = 0.0388; FX[0][45] = -0.0004; FX[1][45] = 0.4709; FX[2][45] = 4.6318; FX[3][45] = 9.9796; FX[4][45] = 7.9628; FX[5][45] = 1.9106; FX[6][45] = 0.0780; FX[7][45] = -0.5627; FX[8][45] = -3.7164; FX[9][45] = -8.9993; FX[10][45] = -8.5153; FX[11][45] = -2.8176; FX[12][45] = -0.3096; FX[13][45] = 0.2406; FX[14][45] = 2.0968; FX[15][45] = 6.9033; FX[16][45] = 9.4641; FX[17][45] = 5.2334; FX[18][45] = 1.0095; FX[19][45] = 0.0104; FX[20][45] = -0.7114; FX[21][45] = -5.5902; FX[22][45] = -10.7380; FX[23][45] = -6.9143; FX[24][45] = -1.1966; FX[25][45] = 0.0849; FX[26][45] = 0.6788; FX[27][45] = 4.2016; FX[28][45] = 9.2459; FX[29][45] = 7.7349; FX[30][45] = 2.7775; FX[31][45] = 0.3947; FX[32][45] = -0.0396; FX[33][45] = -2.3831; FX[34][45] = -8.2531; FX[35][45] = -9.5958; FX[36][45] = -4.3474; FX[37][45] = -0.4220; FX[38][45] = 0.0427; FX[0][46] = 0.0044; FX[1][46] = 0.3305; FX[2][46] = 4.2614; FX[3][46] = 9.8622; FX[4][46] = 8.5410; FX[5][46] = 2.0331; FX[6][46] = -0.0014; FX[7][46] = -0.5653; FX[8][46] = -3.6369; FX[9][46] = -8.8995; FX[10][46] = -8.4828; FX[11][46] = -2.9743; FX[12][46] = -0.4221; FX[13][46] = 0.2740; FX[14][46] = 1.9851; FX[15][46] = 6.7785; FX[16][46] = 9.5642; FX[17][46] = 5.3612; FX[18][46] = 1.0414; FX[19][46] = 0.0132; FX[20][46] = -0.9000; FX[21][46] = -5.5977; FX[22][46] = -10.4684; FX[23][46] = -6.8989; FX[24][46] = -1.2227; FX[25][46] = 0.0814; FX[26][46] = 0.7596; FX[27][46] = 4.6729; FX[28][46]

= 9.1934; FX[29][46] = 7.5627; FX[30][46] = 2.4701; FX[31][46] = 0.3156; FX[32][46] =
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$V[26][7] = 0.9848$; $V[27][7] = 0.7902$; $V[28][7] = 0.1476$; $V[29][7] = -0.6128$; $V[30][7] = -0.9502$; $V[31][7] = -0.9998$; $V[32][7] = -1.0025$; $V[33][7] = -0.9041$; $V[34][7] = -0.3879$; $V[35][7] = 0.4165$; $V[36][7] = 0.8988$; $V[37][7] = 0.9985$; $V[38][7] = 0.9983$; $V[0][8] = 1.0012$; $V[1][8] = 0.9957$; $V[2][8] = 0.7904$; $V[3][8] = 0.0996$; $V[4][8] = -0.6574$; $V[5][8] = -0.9658$; $V[6][8] = -1.0008$; $V[7][8] = -0.9974$; $V[8][8] = -0.8668$; $V[9][8] = -0.3232$; $V[10][8] = 0.4341$; $V[11][8] = 0.8697$; $V[12][8] = 0.9793$; $V[13][8] = 0.9889$; $V[14][8] = 0.9028$; $V[15][8] = 0.4704$; $V[16][8] = -0.3177$; $V[17][8] = -0.8747$; $V[18][8] = -1.0012$; $V[19][8] = -1.0041$; $V[20][8] = -0.9717$; $V[21][8] = -0.6161$; $V[22][8] = 0.1523$; $V[23][8] = 0.7781$; $V[24][8] = 0.9763$; $V[25][8] = 0.9976$; $V[26][8] = 0.9871$; $V[27][8] = 0.7970$; $V[28][8] = 0.1476$; $V[29][8] = -0.6238$; $V[30][8] = -0.9512$; $V[31][8] = -1.0021$; $V[32][8] = -1.0022$; $V[33][8] = -0.9059$; $V[34][8] = -0.3817$; $V[35][8] = 0.4458$; $V[36][8] = 0.9243$; $V[37][8] = 1.0009$; $V[38][8] = 0.9973$; $V[0][9] = 1.0011$; $V[1][9] = 0.9947$; $V[2][9] = 0.7856$; $V[3][9] = 0.0919$; $V[4][9] = -0.6625$; $V[5][9] = -0.9611$; $V[6][9] = -0.9964$; $V[7][9] = -0.9849$; $V[8][9] = -0.8409$; $V[9][9] = -0.3080$; $V[10][9] = 0.4398$; $V[11][9] = 0.8764$; $V[12][9] = 0.9801$; $V[13][9] = 0.9944$; $V[14][9] = 0.9201$; $V[15][9] = 0.4919$; $V[16][9] = -0.3207$; $V[17][9] = -0.8741$; $V[18][9] = -0.9970$; $V[19][9] = -1.0033$; $V[20][9] = -0.9734$; $V[21][9] = -0.6091$; $V[22][9] = 0.1518$; $V[23][9] = 0.7723$; $V[24][9] = 0.9702$; $V[25][9] = 0.9900$; $V[26][9] = 0.9685$; $V[27][9] = 0.7645$; $V[28][9] = 0.1047$; $V[29][9] = -0.6403$; $V[30][9] = -0.9572$; $V[31][9] = -1.0007$; $V[32][9] = -1.0015$; $V[33][9] = -0.8867$; $V[34][9] = -0.3229$; $V[35][9] = 0.4947$; $V[36][9] = 0.9425$; $V[37][9] = 1.0017$; $V[38][9] = 0.9988$; $V[0][10] = 1.0015$; $V[1][10] = 0.9968$; $V[2][10] = 0.7804$; $V[3][10] = 0.0700$; $V[4][10] = -0.6860$; $V[5][10] = -0.9605$; $V[6][10] = -0.9951$; $V[7][10] = -0.9818$; $V[8][10] = -0.8338$; $V[9][10] = -0.2967$; $V[10][10] = 0.4486$; $V[11][10] = 0.8803$; $V[12][10] = 0.9842$; $V[13][10] = 1.0037$; $V[14][10] = 0.9510$; $V[15][10] = 0.5299$; $V[16][10] = -0.3041$; $V[17][10] = -0.8651$; $V[18][10] = -0.9885$; $V[19][10] = -1.0020$; $V[20][10] = -0.9484$; $V[21][10] = -0.5387$; $V[22][10] = 0.2351$; $V[23][10] = 0.8113$; $V[24][10] = 0.9749$; $V[25][10] = 0.9788$; $V[26][10] = 0.9341$; $V[27][10] = 0.6916$; $V[28][10] = 0.0196$;

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